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Attorney's Docket No. 5308-159

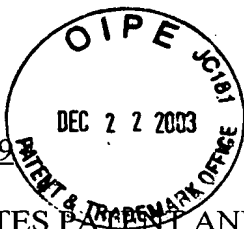


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PATENT

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re: Thomas G. Coleman

Serial No.: 09/931,537

Filed: August 16, 2001

For: SILICON CARBIDE SUBLIMATION SYSTEMS AND ASSOCIATED METHODS

Group: 1765

Examiner: M. Song

Confirmation No. 3269

Mail Stop Appeal-Brief Patents

Commissioner for Patents

P.O. Box 1450

Alexandria, VA 22313-1450

December 17, 2003

**TRANSMITTAL OF APPEAL BRIEF
(PATENT APPLICATION--37 C.F.R. § 1.192)**

1. Transmitted herewith, in triplicate, is the APPEAL BRIEF in this application, with respect to the Notice of Appeal filed on October 20, 2003.
2. This application is filed on behalf of
☐ a small entity
A verified statement ☐ is attached; ☐ was already filed.
3. Pursuant to 37 C.F.R. § 1.17(c), the fee for filing the Appeal Brief is:

<input type="checkbox"/> small entity	\$165.00
<input checked="" type="checkbox"/> other than small entity	\$330.00

Appeal Brief fee due \$330.00

- ☒ The Appeal Brief fee due, and any additional fee or refund may be charged to Deposit Account 50-0220.

Respectfully submitted,

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Susan E. Freedman

Date of Signature: December 17, 2003

Attorney Docket No.: 5308-159

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APPELLANTS' BRIEF ON APPEAL UNDER 37 C.F.R. § 1.192

Sir:

This Appeal Brief is filed in triplicate pursuant to the *Notice of Appeal to the Board of Patent Appeals and Interferences* mailed October 20, 2003.

REAL PARTY IN INTEREST

The real party in interest is assignee Cree, Inc., Durham, North Carolina.

RELATED APPEALS

Appellant is aware of no appeals or interferences that would be affected by the present appeal.

STATUS OF CLAIMS

Claims 1-5, 7-24, 26-33, 35-36 and 46-48 remain pending. Each of these claims currently stands finally rejected. Appellants appeal the final rejection of Claims 1-5, 7-24, 26-33, 35-36 and 46-48. The attached Appendix A presents the claims at issue as finally rejected in the Official Action of July 8, 2003 and the Advisory Action of October 9, 2003.

STATE OF AMENDMENTS

The attached Appendix A presents the claims as they currently stand. Each of the claims remains as filed or as amended in the Amendment dated May 1, 2003. All amendments have been entered in the present case.

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SUMMARY OF THE INVENTION

Pursuant to embodiments of the present invention, methods of growing silicon carbide crystals using electric arc sublimation techniques are provided. (Specification at 5, lines 1-2). Sublimation refers to a process whereby a material is converted from a solid state directly into a gaseous state and thereafter the resulting gas vapors are condensed back into a solid form. (See Webster's New World Dictionary at 1418, Appendix B). In embodiments of the present invention, at least one silicon carbide electrode may be placed in a sublimation system. (Specification at 5, lines 2-8). An electric arc may then be created between the silicon carbide electrode and a second electrode that raises the temperature of the silicon carbide electrode adjacent the arc to a temperature sufficient to sublime the silicon carbide. (Specification at 5, lines 2-8). In other embodiments of the invention, a localized heat source may be used instead of the electric arc to raise at least a portion of the silicon carbide source material to a temperature at which it sublimates. (Specification at 5, lines 8-10).

The power dissipation across the electric arc may be controlled to create a constant flow of vaporized Si, Si₂C and/or SiC₂ from the silicon carbide source independent of both the internal temperature and/or pressure of the sublimation system. (Specification at 5, lines 11-15). This may be accomplished, for example, by adjusting the position of one or both electrodes as the electric arc consumes the end of the electrodes so as to maintain a constant gap between the electrodes over time. (See, e.g., Specification at 10, lines 21-29). By using the power dissipation level across the electric arc as a means for maintaining a constant flux, the temperature and pressure of the sublimation system may be independently set to grow a desired polytype of silicon carbide. (Specification at 11, lines 19-29). This differs from conventional silicon carbide sublimation techniques where the pressure and/or temperature may be adjusted throughout the process in an effort to maintain a relatively constant source-to-seed flux of the vaporized silicon carbide. (Specification at 11, lines 19-29).

ISSUES

1. Are Claims 1-5, 7-12, 14-17, 20, 23-24, 26-33, 35-36 and 46-47 properly rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov (U.S. Patent No.

4,978,556) in view of Davis (U.S. Patent No. RE 34,861) and Kuehnle (U.S. Patent No. 5,879,518)?

2. Are Claims 1-5, 7-12, 14, 17-20, 23-24, 26-30, 32-33, 35-36 and 46-47 properly rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis and Smalley (U.S. Patent No. 5,227,038)?

3. Is Claim 13 properly rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Jaussaud (U.S. Patent No. 6,113,692), and Kuehnle or Smalley?

4. Are Claims 18-19 properly rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Kuehnle and Fey (U.S. Patent No. 4,582,004)?

5. Are Claims 21-22 properly rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Kuehnle or Smalley and Otsuki (U.S. Patent No. 6,090,733)?

6. Are Claims 22 and 48 properly rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Kuehnle or Smalley and Kijima (U.S. Patent No. 5,093,039)?

GROUPING OF CLAIMS

Claims 1-5, 7-24, 26-33, 35-36 and 46-48 stand rejected as obvious under 35 U.S.C. § 103. For the purposes of this appeal, Appellant submits that Claims 1-5, 7-10, 13, 16-24, 26-29, 31-32 and 46-48 may be considered as standing or falling together (Group I), Claims 33, 35 and 36 may be considered as standing or falling together (Group II), Claims 14 and 30 may be considered as standing or falling together (Group III), Claims 11 and 15 may be considered as standing or falling together (Group IV) and Claim 12 may also be considered as standing or falling independently of the remaining claims (Group V). Appellant submits that the above-listed groups of claims are separately patentable for the reasons discussed below.

ARGUMENT

I. INTRODUCTION

Each of the claims of the present application stands rejected as obvious under 35 U.S.C. § 103. A determination under Section 103 that an invention would have been obvious to someone of ordinary skill in the art is a conclusion of law based on fact. *Panduit Corp. v. Dennison Mfg. Co.* 810 F.2d 1593, 1 U.S.P.Q.2d 1593 (Fed. Cir. 1987), *cert. denied*, 107 S.Ct. 2187. After the involved facts are determined, the decision maker must then make the legal determination of whether the claimed invention as a whole would have been obvious to a person having ordinary skill in the art at the time the invention was unknown, and just before it was made. *Id.* at 1596. The United States Patent and Trademark Office has the initial burden under Section 103 to establish a *prima facie* case of obviousness. *In re Fine*, 837 F.2d 1071, 5 U.S.P.Q.2d 1596, 1598 (Fed. Cir. 1988).

To establish a *prima facie* case of obviousness, the prior art references cited in the rejection, when combined, must teach or suggest **all** the recitations of the claims, and there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to combine the reference teachings in the manner suggested. M.P.E.P. § 2143. The mere fact that references can be combined or modified does not render the resultant combination obvious unless the prior art also suggests the desirability of the combination. M.P.E.P. § 2143.01, citing *In re Mills*, 916 F.2d 680, 16 U.S.P.Q.2d 1430 (Fed. Cir. 1990). As emphasized by the Court of Appeals for the Federal Circuit, to support combining references, evidence of a suggestion, teaching, or motivation to combine must be clear and particular, and this requirement for clear and particular evidence is not met by broad and conclusory statements about the teachings of references. *In re Dembiczak*, 50 U.S.P.Q.2d 1614, 1617 (Fed. Cir. 1999). Thus, in support of a Section 103 rejection, particular evidence from the prior art must be provided showing why a skilled artisan, with no knowledge of the claimed invention, would have combined the cited references in the manner claimed in the rejection. *In re Kotzab*, 55 U.S.P.Q.2d 1313, 1317 (Fed. Cir. 2000).

Furthermore, as recently stated by the Federal Circuit with regard to the selection and combination of references:

This factual question of motivation is material to patentability, and could not be resolved on subjective belief and unknown authority. It is improper, in determining whether a person of ordinary skill would have been led to this combination of references, simply to "[use] that which the inventor taught against its teacher." *W.L. Gore v. Garlock, Inc.*, 721 F.2d 1540, 1553, 220 USPQ 303, 312-13 (Fed. Cir. 1983). Thus the Board must not only assure that the requisite findings are made, based on evidence of record, but must also explain the reasoning by which the findings are deemed to support the agency's conclusion....

In re Sang Su Lee, 277 F.3d 1338, 1343 (Fed. Cir. 2002).

Appellant respectfully submits that the pending claims are patentable over the cited references because the cited combination fails to disclose or suggest all of the recitations of the pending claims, and because the reasoning behind such combination has not been established. The patentability of the pending claims is discussed in detail hereinafter.

II. THE GROUP I-V CLAIMS ARE PATENTABLE OVER THE CITED ART

Claims 1-5, 7-12, 15-17, 20, 23-24, 26-29, 31-32 and 46-47 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis and Kuehnle. (Final Action at ¶ 2, pp. 2-5). Claims 1-5, 7-12, 17-20, 23-24, 26-29, 32, and 46-47 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis and Smalley. (Final Action at ¶ 3, pp. 5-7). Claim 13 stands rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Jaussaud, and Kuehnle or Smalley. (Final Action at ¶ 4, pp. 7-8). Claims 18-19 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Kuehnle and Fey. (Final Action at ¶ 5, pp. 8-9). Claims 21-22 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Kuehnle or Smalley and Otsuki. (Final Action at ¶ 6, pp. 9-10). Claims 22 and 48 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Kuehnle or Smalley and Kijima. (Final Action at ¶ 7, pp. 10-11). For the reasons discussed below, Appellant respectfully submits that all of these claims are patentable over the cited art.

A. The Rejections Should Be Reversed Because Pinkahsov Does Not Teach Growing Silicon Carbide Via Sublimation

The present application includes three independent claims, Claims 1, 24 and 33. For the Court's convenience, each of these claims are reproduced below:

1. A method of growing silicon carbide, the method comprising:
introducing a seed of silicon carbide into a sublimation system;
introducing a silicon carbide electrode into the sublimation system;
introducing a second electrode into the sublimation system adjacent the silicon carbide electrode, wherein the silicon carbide electrode and the second electrode are separated by a gap;
establishing an electric arc across the gap between the silicon carbide electrode and the second electrode to vaporize at least part of the silicon carbide electrode and cause at least some of the vaporized silicon carbide materials to form silicon carbide on the silicon carbide seed; and
controlling the power dissipated across the gap to control a flow of vaporized Si, Si₂C and SiC₂ from the silicon carbide electrode to the seed of silicon carbide.

24. A method of growing silicon carbide, the method comprising:
establishing an electrical arc between a pair of electrodes spaced apart by a gap in order to electrically arc a silicon carbide source to sublimate silicon and carbon containing material from the silicon carbide source and cause at least some of the silicon and carbon containing material to form silicon carbide on a silicon carbide seed; and
controlling the power dissipated across the gap to control the flow of vaporized Si, Si₂C and SiC₂ from the silicon carbide source to the silicon carbide seed.

33. A method of growing silicon carbide, the method comprising:
using resistive or inductive heating to heat a furnace to a temperature below the temperature at which silicon carbide sublimates;
using an electric arc to create a local high temperature zone within a the furnace that is above the temperature at which silicon carbide sublimates while maintaining the inner walls of the furnace at a temperature below the temperature at which silicon carbide sublimates;
introducing a silicon carbide source material into the high temperature zone to sublimate silicon and carbon containing material from the silicon carbide source and cause at least some of the silicon and carbon containing material to form silicon carbide on a silicon carbide seed.

As is clear from the above, all of the pending claims are directed to methods of **growing silicon carbide via electric arc sublimation**. (See, e.g., Claim 1, stating that a seed of silicon carbide is introduced "into a sublimation system"; see also Claims 24 and 33, stating that an electrical arc is used "to sublimate silicon and carbon." The Advisory Action states that the Pinkahsov reference is relied upon as teaching the electric arc sublimation of silicon carbide recitations of the pending claims. (See Advisory Action at 2).

Appellant respectfully submits that the rejections of all of the pending claims should be withdrawn because Pinkahsov does not teach sublimating silicon carbide using electric arc vapor deposition techniques. Sublimation refers to a process where a solid is heated directly to a gaseous state and the resulting vapors are condensed back into solid form. (See Webster's Dictionary at 1418, defining sublimate and sublime, attached as Appendix B hereto). Pinkahsov, however, is directed to an arc vapor deposition technique in which molten (*i.e.*, material that has been "melted or liquefied by heat") material is vaporized for purposes of growing a material on a substrate. (See Pinkahsov at Col. 2, lines 34-36; Col. 3, lines 49-54; *see also* Webster's Dictionary at 916, Appendix B, defining molten). Evaporating a molten liquid, by definition, is not sublimation. Thus, as none of the cited references teach or suggest growing silicon carbide via electric arc sublimation the rejections should be reversed.

In the Advisory Action, the Examiner argues that the Pinkahsov reference in fact teaches sublimation and is not limited to embodiments that "require a molten pool." (See Advisory Action at 2, citing to Pinkahsov at Col. 1, lines 34-43 and Claim 5). However, the portion of Pinkahsov cited by the Examiner is a summary of arc-vapor deposition techniques that are disclosed in a series of prior patents to Pinkahsov. (See Pinkahsov at Col. 1, lines 29-43). Pinkahsov expressly states that it is these prior patents that describe the details regarding "[t]he formation of vapor and its deposition upon the substrate." (Pinkahsov at Col. 3, lines 54-57). These prior patents to Pinkahsov make clear that the Pinkahsov process involves two phase transformations – solid to liquid and liquid to gas.

For instance, one of the prior patents that is summarized in Pinkahsov describes the arc vapor deposition technique as follows:

[A] body of the material to be transferred to the substrate is heated in the region of this substrate and transformed first into a molten state and then into a vapor state. The material thus undergoes two phase transformations, namely, the transformation from the solid phase to the liquid phase and then from the liquid phase to the vapor phase.

(U.S. Patent No. 4,505,948 to Pinkahsov at Col. 1, lines 42-48). The prior patents go on to explain that the vapor deposition proceeds by striking an electric arc between the pool of molten material and an electrode:

This application discloses a method of vapor-depositing material upon a substrate which, as indicated, utilizes an electrical arc struck between a pool of molten

material and a counterelectrode, thereby vaporizing the material on the surface of the pool and permitting transfer of the vaporized material in the vapor state to the substrate.

(*Id.* at Col. 2, lines 13-17). The Pinkahsov patent cited by the Examiner likewise confirms that it relates to a process in which molten materials are vaporized by repeatedly stating that the arc is applied to a **molten electrode material**. (Pinkahsov at Col. 2, lines 31-36 and Col. 3, lines 49-54).

In summary, the portions of Pinkahsov relied upon by the Examiner to support the pending rejections do not teach growing silicon carbide via electric arc **sublimation** as recited in all of the pending claims, but instead disclose a two-step vaporization process that does not involve sublimation. The Examiner cannot change this fact by citing to sentences in Pinkahsov that describe the vaporization process in summary fashion as a teaching that sublimation could be used instead of a two-step process. As such, the rejections of all the pending claims should be reversed.¹

B. Pinkahsov Does Not Enable Arc-Vapor Deposition Using a Silicon Carbide Electrode

The rejections of the pending claims should likewise be reversed because Pinkahsov does not **enable** a person of skill in the art to perform electric arc vapor deposition using a silicon carbide electrode. As noted above, Pinkahsov discloses an arc vapor-deposition method in which an arc is struck between a molten pool of electrode material and a second electrode. Pinkahsov states that "the electrode material is crystalline silicon or silicon carbide or silicon nitride." (Pinkahsov at Col. 2, lines 47-48). However, the example in Pinkahsov only describes use of two **silicon** electrodes. Pinkahsov does further state that the manner in which the vapor is formed is fully described in a series of

¹ The Advisory Action also states that "Smalley et al teaches a method of electric arc processing, where electrode material is vaporized without forming a molten pool." (Advisory Action at 2). Smalley, however, relates to an electric arc process that uses a different material system for a completely different purpose, namely generating fullerenes by vaporizing carbon. (See Smalley at Abstract). The Examiner has not even attempted to explain why a skilled artisan would have been motivated to modify the process of Pinkahsov to operate via sublimation based on Smalley, and Appellant respectfully submits that the only such motivation comes from hindsight and using the present invention as a roadmap to piece together various teachings from a wide variety of prior art references.

prior, related patents. (Pinkahsov at Col. 1, lines 29-34 and Col. 3, lines 54-57). These prior patents, however, explain that the silicon carbide is deposited on the substrate by using a silicon electrode and a carbon block electrode which contains "a molten pool of silicon and solubilized carbon." (See U.S. Patent No. 4,505,948 to Pinkahsov at Col. 6, lines 60-63 and Col. 7, lines 36-40). Thus, Pinkahsov does not explain how the process could be carried out using a silicon carbide electrode.

This failure of Pinkahsov is significant because it is clear that the process of Pinkahsov **would not work** with a silicon carbide electrode. Pinkahsov teaches that the process is carried out under vacuum conditions (*i.e.*, at pressures of 10^{-3} torr or better). (Pinkahsov at Col. 3, lines 66-68). As shown in the references attached at Appendix C hereto, silicon carbide **does not form a liquid state at those pressures** (See, *e.g.*, Seace and Slack, Solubility of Carbon in Silicon and Germanium at 1554). Accordingly, the method of Pinkahsov where the arc vapor deposition is carried out between an electrode and a pool of molten material in a second electrode will not work with silicon carbide electrodes. Thus, because Pinkahsov does not enable an arc vapor-deposition process that uses a silicon carbide electrode, the rejections of all the pending claims should be withdrawn.

C. The Rejections Should Be Reversed Because There is No Motivation To Combine Kuehnle or Smalley with Pinkahsov and Davis

As noted above, to establish a *prima facie* case of obviousness, the combination of prior art references must teach or suggest all the recitations of the claims **and** there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to combine the reference teachings to arrive at the claimed invention. M.P.E.P. § 2143. The mere fact that references can be combined or modified does not render the resultant combination obvious unless the prior art also suggests the desirability of the combination.

All of the pending claims stand rejected under 35 U.S.C. § 103 based, at least in part, on (1) the combination of Pinkahsov, Davis and Kuehnle and (2) the combination of Pinkahsov, Davis and Smalley. In these rejections, the Examiner relies on Kuehnle or Smalley for teaching the step of "controlling the power dissipated across the gap to control

a flow of vaporized Si, Si₂C and SiC₂ from the silicon carbide electrode to the seed of silicon carbide." (*See, e.g.*, Final Office Action at ¶ 2, p. 4). The Final Office Action, however, fails to provide the necessary showing that a skilled artisan would have been motivated to combine this alleged teaching of either Kuehnle or Smalley with Pinkahsov and Davis. The lack of any such evidence of motivation to combine the references in the manner of the rejections provides an independent basis for withdrawal of each of the pending rejections.

The portion of the Final Office Action detailing the rationale for the pending rejections conclusively states that "[i]t would have been obvious to a person of ordinary skill in the art at the time of the invention to modify the combination of Pinkahsov and Davis et al with Kuehnle to vaporize the electrode material at a steady level (col. 4, ln 15-25), thereby avoiding undesired changes in flux resulting in uniform polytype." (Final Office Action at 4). With respect to the rejections that rely on Smalley, the Final Office Action similarly states in a conclusory fashion that "[i]t would have been obvious to a person of ordinary skill in the art at the time of the invention to modify the combination of Pinkahsov and Davis et al with Smalley's means for maintaining a desired gap for the electrical arc to maintain an optimum length of the arc gap during the entire process." (Final Office Action at 6). In the Advisory Action, the Examiner argues that the teaching in Davis that the silicon carbide should be vaporized at a constant flux provides the motivation for combining Kuehnle and/or Smalley with Pinkahsov and Davis. (Advisory Action at 3). However, Appellant respectfully submits that a skilled artisan would not combine the references as suggested in the pending rejections for at least two independent reasons.

First, neither Kuehnle nor Smalley relate to depositing silicon carbide on a substrate via electric arc sublimation. Kuehnle is directed to evaporating particles at a steady state so that the particles will cluster together at a consistent rate. (*See* Kuehnle at Col. 4, lines 13-23). Smalley is directed to a method of forming fullerenes, a molecular form of carbon. (*See* Smalley at Abstract). Thus, the suggestions in both Kuehnle and Smalley regarding the gap to maintain between the electrodes is made in the context of a different art and directed to solving a different problem. The **Davis reference, however, contains explicit teachings regarding controlling the flux between the silicon carbide**

source material and the silicon carbide seed. In particular, Davis teaches that the "if the thermal gradient is continually increased as the source powder is depleted and as the seed crystal grows, an absolute temperature differential between the source and the seed can be maintained at an amount which continues to be most favorable for growth." (Davis at Col. 9, lines 29-35). Davis further teaches that this can be accomplished by modifying the temperature of the source powder and/or the seed during growth and/or by changing the pressure during growth or by a combination of these methods. Appellant respectfully submits that the skilled artisan would look to the teachings in the Davis references regarding controlling the temperature and the pressure in the reactor to maintain a steady flux or the teachings of some other reference that actually relates to the field of the present invention. What the skilled artisan would not do is look to a completely different field to directly modify the teachings of a reference that the Final Office Action contends teaches the method of solving the problem addressed by the present invention. Thus, the pending rejections should be withdrawn because the Examiner has not and cannot show that a skilled artisan would have combined Pinkashov and Davis with either Kuehnle or Smalley in the manner suggested.

The combinations of references relied upon in the rejections is also unsupportable because **Pinkahsov teaches directly away from using the electrode apparatus disclosed in Kuehnle or Smalley.** In particular, Pinkahsov teaches that "arc vapor deposition" of silicon or silicon carbide onto a substrate is accomplished by repeatedly striking two electrodes together and then separating the electrodes to form an arc between them. (Pinkahsov at Col. 1, lines 34-45 and Co. 3, lines 49-60). In the example set forth in Pinkahsov, an arc is formed approximately one time per second through this intermittent movement of one or both of the electrodes. (Pinkahsov at Col. 4, lines 27-28). As explained in one of the earlier patents that Pinkahsov references as describing aspects of his invention:

I may move the counterelectrode into and out of contact with the pool to thereby deposit some of the melt upon the counterelectrode and permitting the heat generated at the electrode tip to vaporize at least in part the material transferred to it and thus in part generate the vapors which are to be transferred to the substrate.

(U.S. Patent No. 5,505,948 at Col. 2, lines 30-36). This prior Pinkahsov patent goes on to state:

Surprisingly, once the arc is struck as the two electrodes are separated, the arc, a portion of the arc or a heating effect generated by the arc appears to spiral around the long electrode and cause vaporization of the material of the electrode in a generally helical or spiral pattern progressively moving away from the counterelectrode.

It is indeed a remarkable surprise that the arc is not confined to the space between the two electrodes but rather has a component or an effect which spirals away from the counterelectrode toward a region of the length of the long electrode which is further removed from the counterelectrode in spite of the fact that the greatest conductivity would appear to lie in a line directly between the two electrodes where the major portion of the arc appears to be confined. This effect is manifest in the fact the long electrode, i.e. the deposition electrode, while originally of uniform cross section, develops a taper toward the counterelectrode and coating from the blank of the deposition electrode onto the substrate can be observed at considerable distance from the arc's striking face of the deposition electrode.

In fact, the effect appears to survive for a brief period following extinction of the original arc and hence I prefer to periodically contact and separate the electrodes to generate the arc and then allow extinction thereof.

(U.S. Patent No. 5,505,948 at Col. 4, lines 16-42). Thus, it is clear that a critical part of the invention of Pinkahsov involves moving the electrodes into and out of contact with each other – in other words, **not** maintaining a constant gap – as this is required both to (1) facilitate vaporization of the electrode material in the molten pool by depositing that material onto the tip of the counterelectrode and (2) to induce an effect whereby the arc is formed not only between the two electrodes, but also along the length of the deposition electrode.

In light of these teachings in Pinkahsov, Appellant respectfully submits that a skilled artisan would not have been motivated to combine either Kuehnle or Smalley with Pinkahsov as doing so would be **directly contrary** to the teachings of Pinkahsov. Appellant notes that in the Advisory Action the Examiner argues that Pinkahsov is not limited to embodiments in which the electrodes are struck together to form the arc. However, what the Examiner has not and cannot do is point to any other arc vaporization process in Pinkahsov, because the only process disclosed is one in which the electrodes are struck together. In fact, the very portion of Pinkahsov that the Examiner cites to as teaching that Pinkahsov is not limited to embodiments in which the electrodes are struck together expressly states that the electrodes are brought into contact and separated to form

the arc. (*See* Advisory Action at 3-4, citing to Pinkahsov at Col. 1, lines 29-45). As such, Appellant respectfully submits that this teaching away from the combination of references cited in the pending rejections compels reversal of those rejections.

D. The Rejections Should Be Withdrawn Because Kuehnle and Smalley are Not Analogous Art

Finally, the pending rejections of the Group I-V claims should also be withdrawn because neither Kuehnle nor Smalley can properly be relied upon as part of a rejection under 35 U.S.C. § 103. In particular, only references that come from an "analogous art" may be relied upon to support a rejection under Section 103, where a reference is considered to be from an analogous art if it either (1) is from the same field of endeavor as the invention at issue, regardless of the problem addressed or (2) is reasonably pertinent to the particular problems with which the inventor is involved. *See, e.g., In re Paulsen*, 31 U.S.P.Q.2d 1671, 1675-76 (Fed. Cir. 1994). Neither Kuehnle nor Smalley satisfy this test.

As to the first prong of the analogous art test, the present invention is directed to the growth of semiconductor materials on a seed crystal or substrate. In contrast, Kuehnle is directed to "a method for producing small particles, e.g., nanoparticles, which have consistent size, shape, structure and functionality." (Kuehnle at Col. 1, lines 7-10). Smalley likewise has nothing to do with growing semiconductor materials on a substrate, but instead is directed to a method for making fullerenes (a particular form of carbon). (*See* Smalley at Abstract). Thus, both Kuehnle and Smalley are from different fields of endeavor, and hence neither reference qualifies as analogous art under the first prong of the test.

The second prong of the analogous art test examines whether or not a reference is reasonably pertinent to the particular problems with which the invention is involved. Here, that problem is growing monocrystalline and/or polycrystalline silicon carbide crystals on a substrate for use in semiconductor applications. Kuehnle has nothing to do with the problems which the present invention addresses, as Kuehnle provides insights regarding how to collect a powder of a source substance where the particles of the powder are relatively uniform in size. Smalley is directed to a method of growing fullerenes – a form of carbon – that has no semiconductor applications. Thus, neither Kuehnle nor Smalley qualify as analogous art under the second prong of the test and, as such, the

claims of the present application cannot properly be rejected based on the combination of the Pinkashov and Davis and either Kuehnle or Smalley.

III. THE GROUP II CLAIMS ARE PATENTABLE OVER THE CITED ART

The Group II claims are Claims 33, 35 and 36. The Group II claims stand rejected under 35 U.S.C. § 103 based on the combination of (1) Pinkahsov, Davis and Kuehnle and (2) Pinkahsov, Davis and Smalley. (*See* Final Office Action at ¶¶ 2, 3, pp. 2-7). Appellant respectfully submits that each of the Group II claims are patentable over the cited references for each of the reasons set forth in Section II above. In addition, Appellant submits that the Group II claims are also patentable over the cited art for at least one additional reason.

Each of the Group II claims recite that the electric arc is used to "create a local high temperature zone within the furnace . . . while maintaining the inner walls of the furnace at a temperature below the temperature at which silicon carbide sublimates." Appellant respectfully submits that none of the cited references teach or suggest "using resistive or inductive heating to heat a furnace" while at the same time "maintaining the inner walls of the furnace at a temperature below the temperature at which silicon carbide sublimates." (*See* Claim 33). Appellant raised the above argument in Appellant's Response to Final Office Action. In response, the Examiner argued in the Advisory Action that the Group II claims do "not require heating the inner walls of the furnace." (Advisory Action at 4). The Examiner, however, **apparently overlooked** the recitation of Claim 33 that recites "using resistive or inductive heating to heat a furnace to a temperature below the temperature at which silicon carbide sublimates." Heating the furnace includes heating the inner walls of the furnace. Thus, as it is clear that the rejections of the Group II claims are based on a mistake by the Examiner regarding the actual elements included within the Group II claims, Appellant respectfully submits that the rejections of the Group II claims should be reversed.

In any event, Appellant submits that:

- Davis teaches heating the furnace walls to a temperature **above** the temperature at which silicon carbide sublimates. (*See, e.g.*, Davis at Col. 11, lines 20-23).

- Pinkahsov does not teach or suggest that a furnace is even provided, as the electric arc is used solely to perform the vaporization. (*See* Pinkahsov at Col. 4, lines 5-41).
- Smalley does not teach or suggest that a furnace is even provided, as the electric arc is used solely to perform the vaporization. (*See* Smalley at Col. 2, lines 46-48).
- Kuehnle teaches that a "cold" inert gas is flowed just inside the housing sidewall, indicating that in Kuehnle the housing is actually cooled as opposed to heated. (*See* Kuehnle at Col. 4, lines 3-12).

This showing that the cited art does not disclose each of the recitations of the Group II claims is un rebutted. Accordingly, Appellant respectfully requests that the rejections of the Group II claims be reversed for this additional reason.

IV. THE GROUP III CLAIMS ARE PATENTABLE OVER THE CITED ART

The Group III claims are Claims 14 and 30. Appellant respectfully submits that each of the Group III claims are patentable over the cited references for the reasons set forth in Section II above. In addition, Appellant submits that the Group III claims are also patentable over the cited art for at least one additional reason.

Claim 14 recites that "the internal temperature of the sublimation system, the position of the silicon carbide electrode and the second electrode, a voltage drop across the gap and a current conducted across the gap are configured so as to maintain the end of the silicon carbide electrode adjacent the gap at a substantially constant temperature during the sublimation process." Claim 30 contains a similar recitation. While the Final Office Action states that the combination of Pinkahsov and Davis in view of either Kuehnle or Smalley teaches these recitations, Appellant respectfully disagrees. Pinkahsov discloses a process where the temperature at the end of the electrode will vary (*i.e.*, not remain constant), and Davis teaches that the temperature of the silicon carbide source material is varied throughout the sublimation process. Kuehnle and Smalley are silent regarding the temperature at the end of the electrode, and hence do not provide the necessary teaching. In any event, even if Kuehnle or Smalley taught the recitations of the Group III claims, no motivation has been identified for modifying the primary references – each of which teach away from maintaining the ends of the electrodes at a constant temperature – in this

manner. Accordingly, the Group III claims likewise are independently patentable over the cited art for at least these additional reasons.

In the Advisory Action, the Examiner takes the position that the recitations of Claims 14 and 30 are "recitation[s] of the intended use of the claimed invention," and hence cannot patentably distinguish the invention from the prior art. (Advisory Action at 5). However, the recitations at issue simply are not recitations of the intended use of the invention, but instead comprise one of the conditions that must be met to practice the claimed methods. Thus, as each of the recitations of the Group III claims are not taught or suggested by the cited references, the rejections of the Group III claims should be reversed.

V. THE GROUP IV CLAIMS ARE PATENTABLE OVER THE CITED ART

The Group IV claims are Claims 11 and 15. Appellant respectfully submits that each of the Group IV claims are patentable over the cited references for the reasons set forth in Section II above. In addition, Appellant submits that the Group III claims are also patentable over the cited art for at least one additional reason.

Each of the Group IV claims recite that a "substantially constant pressure level is pre-selected for growth of a pre-selected polytype of silicon carbide." The Advisory Action takes the position that Davis teaches this recitation at Column 13, lines 1-25. Appellant respectfully submits that this is not the case. Specifically, the cited portion of Davis states that the furnace was initially maintained at a pressure of 400 Torr during the growth process. Then, over a period of 85 minutes, the system was slowly evacuated to reduce the pressure from 400 Torr to 10 Torr. The pressure remained at 10 Torr for 6 hours, and then the pressure was increased to 760 Torr. Thus, the cited portion of Davis teaches that the silicon carbide was grown under a variety of different pressure conditions, and clearly does not teach or suggest pre-selecting a pressure level for growth of a pre-selected polytype of silicon carbide. Accordingly, the rejections of the Group IV claims should also be reversed.

VI. THE GROUP V CLAIM IS PATENTABLE OVER THE CITED ART

The Group V claim is Claim 12. Appellant respectfully submits that Claim 12 is patentable over the cited references for the reasons set forth in Section II above. In addition,

Appellant submits that Claim 12 is also patentable over the cited art for at least one additional reason.

Claim 12 recites "raising the temperature of the silicon carbide electrode to a temperature lower than the temperature at which silicon carbide sublimes." Neither the Final Office Action nor the Advisory Action even attempt to explain where this recitation may be found in the cited references. Davis clearly does not teach it, as Davis does not disclose an electrode and, in any event, teaches heating the silicon carbide source material to a temperature above the temperature at which silicon carbide sublimes. Pinkashov, Kuehnle and Smalley do not teach raising the temperature of the silicon carbide electrode at all, aside from the temperature increase that is caused by the electric arc vaporization, and that temperature increase is, again, to a temperature above the temperature at which silicon carbide sublimes. Accordingly, Claim 12 is also independently patentable over the cited art for at least these additional reasons.

V. CONCLUSION

In light of the above discussion, Appellant submits that each of the pending claims is patentable over the cited references and, therefore, request reversal of the rejections of Claims 1-5, 7-24, 26-33, 35-36 and 46-48.

Respectfully submitted,



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CERTIFICATE OF MAILING

I hereby certify that this correspondence is being deposited with the United States Postal Service as first class mail in an envelope addressed to: Mail Stop Appeal-Brief Patents, Commissioner for Patents, P.O. Box 1450, Alexandria, VA, 22313-1450, on December 17, 2003.



Susan E. Freedman

Date of Signature: December 17, 2003

APPENDIX A
Pending Claims USSN 09/931,537
Filed August 16, 2001

1. (Previously Amended): A method of growing silicon carbide, the method comprising:

introducing a seed of silicon carbide into a sublimation system;

introducing a silicon carbide electrode into the sublimation system;

introducing a second electrode into the sublimation system adjacent the silicon carbide electrode, wherein the silicon carbide electrode and the second electrode are separated by a gap;

establishing an electric arc across the gap between the silicon carbide electrode and the second electrode to vaporize at least part of the silicon carbide electrode and cause at least some of the vaporized silicon carbide materials to form silicon carbide on the silicon carbide seed; and

controlling the power dissipated across the gap to control a flow of vaporized Si, Si₂C and SiC₂ from the silicon carbide electrode to the seed of silicon carbide.

2. (Original): The method of Claim 1, wherein the second electrode is a silicon carbide electrode.

3. (Previously Amended): The method of Claim 2, wherein controlling the power dissipated across the gap further controls a flow of vaporized Si, Si₂C and SiC₂ from the second electrode to the seed of silicon carbide.

4. (Previously Amended): The method of Claim 3, wherein controlling the power dissipated across the gap comprises moving at least one of the silicon carbide electrode and the second electrode as they vaporize during the sublimation process to maintain a constant gap between the silicon carbide electrode and the second electrode.

5. (Original): The method of Claim 2, further comprising maintaining the pressure within the sublimation system at a substantially constant level during the sublimation process.

6. (Cancelled)

7. (Previously Amended): The method of Claim 1, wherein the power dissipated across the gap is controlled to maintain a substantially constant flow of vaporized Si, Si₂C and SiC₂ per unit area per unit time from the silicon carbide electrode to the seed crystal.

8. (Previously Amended): The method of Claim 1, wherein controlling the power dissipated across the gap comprises moving at least one of the silicon carbide electrode and the second electrode during the sublimation process to maintain a constant gap between the silicon carbide electrode and the second electrode.

9. (Original): The method of Claim 1, further comprising moving at least one of the silicon carbide electrode and the second electrode to maintain a substantially constant separation between the silicon carbide electrode and the second electrode.

10. (Original): The method of Claim 1, further comprising maintaining the pressure within the sublimation system at a substantially constant level during the sublimation process.

11. (Previously Amended): The method of Claim 10, where the substantially constant pressure level is set to ensure that a specific polytype of silicon carbide is grown.

12. (Original): The method of Claim 10, further comprising:
raising the temperature of the seed to a temperature lower than the temperature at which silicon carbide sublimes; and
raising the temperature of the silicon carbide electrode to a temperature lower than the temperature at which silicon carbide sublimes.

13. (Previously Amended): The method of Claim 12, wherein the sublimation system includes a furnace, and wherein the method further comprises raising the temperature of the inner walls of the furnace to a temperature higher than the temperature of the seed.

14. (Previously Amended): The method of Claim 1, wherein the internal temperature of the sublimation system, the position of the silicon carbide electrode and the second electrode, a voltage drop across the gap and a current conducted across the gap are configured

so as to maintain the end of the silicon carbide electrode adjacent the gap at a substantially constant temperature during the sublimation process.

15. (Previously Amended): The method of Claim 14, where the substantially constant temperature is set to ensure that a specific polytype of silicon carbide is grown.

16. (Previously Amended): The method of Claim 1, wherein controlling the power dissipated across the gap to control a flow of vaporized Si, Si₂C and SiC₂ from the silicon carbide electrode to the seed crystal comprises:

sensing a voltage drop across the gap; and

adjusting the relative location of the silicon carbide electrode and the second electrode so as to maintain the voltage drop at a constant level.

17. (Original): The method of Claim 16, further comprising rotating the seed during at least part of the sublimation process.

18. (Previously Amended): The method of Claim 1, wherein establishing an electric arc between the silicon carbide electrode and the second electrode comprises activating an alternating current power supply that is electrically connected to one of the silicon carbide electrode and the second electrode.

19. (Original): The method of Claim 18, wherein the frequency at which the alternating current power supply is operated is selected to maintain substantially the same rate of vaporization of the silicon carbide electrode.

20. (Original): The method of Claim 2, wherein the silicon carbide electrode is formed by sintering silicon carbide powder.

21. (Original): The method of Claim 20, wherein the silicon carbide electrode is formed from an n-type carrier rich silicon carbide source powder.

22. (Original): The method of Claim 20, wherein the silicon carbide electrode is formed from a p-type carrier rich silicon carbide source powder.

23. (Previously Amended): The method of Claim 2, wherein the internal temperature of the sublimation system, the pressure within the sublimation system and the voltage and current associated with the electric arc are maintained so as to heat a constant volume of the silicon carbide electrode above the temperature where sublimation occurs during a crystal growth phase of the sublimation process.

24. (Previously Amended): A method of growing silicon carbide, the method comprising:

establishing an electrical arc between a pair of electrodes spaced apart by a gap in order to electrically arc a silicon carbide source to sublimate silicon and carbon containing material from the silicon carbide source and cause at least some of the silicon and carbon containing material to form silicon carbide on a silicon carbide seed; and

controlling the power dissipated across the gap to control the flow of vaporized Si, Si₂C and SiC₂ from the silicon carbide source to the silicon carbide seed.

25. (Cancelled)

26. (Previously Amended): The method of Claim 24, wherein the electrodes are silicon carbide electrodes that serve as the silicon carbide source.

27. (Original): The method of Claim 26, wherein the power dissipated across the gap is controlled to maintain a substantially constant flow of vaporized Si, Si₂C and SiC₂ per unit area per unit time from the pair of silicon carbide electrodes to the silicon carbide seed.

28. (Original): The method of Claim 26, wherein controlling the power dissipated across the gap comprises moving at least one of the pair of silicon carbide electrodes as they vaporize during the sublimation process to maintain a constant gap between the pair of silicon carbide electrodes.

29. (Original): The method of Claim 24, further comprising maintaining the pressure within the sublimation system at a substantially constant level during the sublimation process.

30. (Previously Amended): The method of Claim 24, wherein the sublimation process occurs within a heated furnace, and wherein internal temperature of the furnace, the position of the pair of silicon carbide electrodes, the voltage drop across the spacing between the pair of electrodes and the arc current are configured so as to maintain the ends of the pair of electrodes adjacent the arc at a substantially constant temperature during the sublimation process.

31. (Original): The method of Claim 26, wherein controlling the power dissipated across the gap to control the flow of vaporized Si, Si₂C and SiC₂ from the pair of silicon carbide electrodes to the silicon carbide seed comprises:

sensing a voltage drop across the gap; and
adjusting the relative location of the silicon carbide electrodes so as to maintain the voltage drop at a constant level.

32. (Previously Amended): The method of Claim 24, wherein the sublimation process occurs within a heated furnace, and wherein an internal temperature of the furnace, a pressure within the furnace and the voltage and current associated with the arc are maintained so as to heat a constant volume of the silicon carbide source above the temperature where sublimation occurs during a the crystal growth phase of the sublimation process.

33. (Previously Amended): A method of growing silicon carbide, the method comprising:

using resistive or inductive heating to heat a furnace to a temperature below the temperature at which silicon carbide sublimates;

using an electric arc to create a local high temperature zone within a the furnace that is above the temperature at which silicon carbide sublimates while maintaining the inner walls of the furnace at a temperature below the temperature at which silicon carbide sublimates;

introducing a silicon carbide source material into the high temperature zone to sublimate silicon and carbon containing material from the silicon carbide source and cause at

least some of the silicon and carbon containing material to form silicon carbide on a silicon carbide seed.

34. (Cancelled)

35. (Original): The method of Claim 33, wherein the silicon carbide source material is introduced into the high temperature zone by moving the silicon carbide source material.

36. (Previously Amended): The method of Claim 33, wherein the silicon carbide source material is introduced into the high temperature zone by moving a heating source used to create the local high temperature zone.

37. (Cancelled)

38. (Cancelled)

39. (Cancelled)

40. (Cancelled)

41. (Cancelled)

42. (Cancelled)

43. (Cancelled)

44. (Cancelled)

45. (Cancelled)

46. (Original): The method of Claim 1, wherein the silicon carbide seed is a monocrystalline seed of silicon carbide, and wherein the silicon carbide formed on the monocrystalline silicon carbide seed is monocrystalline silicon carbide.

47. (Original): The method of Claim 24, wherein the silicon carbide formed on the silicon carbide seed is monocrystalline silicon carbide.

48. (Original): The method of Claim 2, wherein the silicon carbide electrode is formed from silicon carbide powder grown by a chemical vapor deposition technique.

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< L. *molestare* < *molestus*, troublesome < *moles*, a burden: see **MOLÉ** 1. to annoy, interfere with, or meddle with so as to trouble or harm, or with intent to trouble or harm 2. to make improper advances to, esp. of a sexual nature — *moles-ta-tion* (mō'les tā'shan, māl'as) *n.* — *mo-lest-er* *n.*

Mo-lière (mōl yer'; mō'lē er'; Fr. mō-lyer') (pseud. of Jean Baptiste Poquelin) 1622–73; Fr. dramatist

Mo-line (mō lēn') [*<* Sp. *molino*; mill] city in NW Ill., on the Mississippi; pop. 46,000

mo-line (mō lin', mō'lin) *adj.* [*<* AngloFr. **moliné* < OFr. *molin*, a mill < VL. **molinum*, for LL. *molina*, MILL: from its resemblance to the iron support for the upper millstone] designating a cross with each arm forked and curved back at the end

Moll (māl) a feminine name: see **MARY** — *n.* [usually m-] [Slang] 1. a gangster's mistress 2. a prostitute

moll-lah (māl'ā) *n.* same as **MULLAH**

mol-les-cent (mō les'nt) *adj.* [L. *molliscens*, prp. of *mollere*, to soften < *mollere*, to be soft < *mollis*, soft: see **MOLLIFY**] softening or tending to soften — *mol-les-cence* *n.*

mol-li-fy (māl'ē fī') *vt.* -fied', -fy'ing [ME. *molliſen*, MFr. *molliſier* < LL. *mollificare*, to soften < L. *mollis*, soft (< IE. **mēdu-*, soft < base **mel-*, to crush, whence **MILL**) + *facere*, to make, do] 1. to soothe the temper of; pacify; appease 2. to make less intense, severe, or violent — **SYN.** see **PACIFY** — *mol'li-fa-tion* *n.* — *mol'li-f'er* *n.*

mol-lusc (māl'ask) *n.* same as **MOLLUSK** — *mol-lus-can* (mō lus'kən) *adj.* *n.*

mol-lus-coid (mō lus'koid) *adj.* 1. of or like a mollusk or mollusks 2. in some classifications, designating or of a group (Molluscoidea) of phyla comprising the brachiopods, phoronids, and bryozoans; all of which have a lophophore — *n.* a molluscoid animal

mol-lusk (māl'ask) *n.* [Fr. *mollusque* < ModL. *Mollusca*, coined by Cuvier < L. *mollusca*, a soft-shelled nut < *molluscus*, soft < *mollis*: see **MOLLIFY**] any of a large phylum (Mollusca) of invertebrate animals including the chitons, oysters, clams, mussels, snails, whelks, slugs, squids, octopuses, etc., characterized by a soft, usually unsegmented body, often enclosed wholly or in part in a mantle and a calcareous shell, and usually having gills and a foot — *mol-lus-kan* (mō lus'kən) *adj.* *n.*

Moll-weide (1774–1825), G. mathematician [an equal-area map projection with the whole earth on one map, showing the prime meridian and all parallels of latitude as straight lines and all other meridians as increasing in curvature toward the margins]

Mol-ly (māl'ē) a feminine name: see **MARY**

mol-ly (māl'ē) *n.*, *pl.* -lies [short for ModL. *Molliensis*, genus name < F. N. *Mollien* (1758–1850), Fr. statesman] any of a genus (*Molliensis*) of various brightly colored tropical and subtropical American fishes often kept in aquariums: also *mol'lie*

mol-ly-cod-dle (māl'ē kād'li) *n.* [**MOLLY** + **CODDLE**] a man or boy used to being coddled, or protected, pampered, etc.: *milk-sop* — *vt.* -dled, -dling to pamper; coddle — *mol'ly-cod'dler* *n.*

Molly Ma-guires (mō gwīrz') [its members were sometimes disguised as women] 1. a secret society organized in Ireland in 1843 to prevent evictions by terrorizing agents of landlords 2. a secret society of Irish-American miners in E Pennsylvania (c. 1865–1875), which opposed oppressive industrial and social conditions, sometimes with physical force

Mol-nár (mōl'nār), Fe-renc (fe'rents) 1878–1952; Hung. playwright & novelist, later in the U.S.

Mo-loch (mō'lāk, māl'ak) same as **MOLECH** — *n.* [m-] a spiny-headed Australian lizard (*Moloch horridus*)

Mo-lo-kai (mō'lō kī') [Haw. < ?] island of Hawaii, southeast of Oahu: site of a leper colony; 259 sq. mi.; pop. 6,000

Mo-lo-tov (mō'lō tāf), V(yacheslav) M(ikhaïlovich) (born Vyacheslav Mikhailovich Skriabin) 1890– Russ. statesman; foreign minister of the U.S.S.R. (1939–49; 1953–56)

Molotov cocktail [after prec.] [Slang] a bottle filled with gasoline, etc. and wrapped in a saturated rag or plugged with a wick, ignited, and hurled as an antitank grenade

molt (mōlt) *vi.* [ME. *mouten* (with unhistoric -l- after **FAULT**, in which the letter was orig. silent) < OE. (*bē*)-*mutian*, to exchange < L. *mutare*, to change: see **MUTATE**] to cast off or shed the hair, outer skin, horns, or feathers at certain intervals, prior to replacement of the castoff parts by a new growth: said of certain animals, as reptiles, birds, etc. — *vt.* to replace by molting — *n.* 1. the act or process of molting 2. the parts so shed — *molt'er* *n.*

mol-ten (mōlt'n) [ME.] archaic *pp.* of **MELT** — *adj.* 1. melted or liquefied by heat 2. made by being melted and cast in a mold

Molt-ke (mōlt'kē) 1. Count Hel-muth (Johannes Ludwig) von (hel'mōt'fōn), 1848–1916; Ger. general 2. Count Hel-muth (Karl Bernhard) von, 1800–91; Ger. field marshal; uncle of prec.

mol-to (mōlt'ō) *adv.* [It. < L. *multum*, much] *Musical* very; much; used in musical directions

Mo-luc-cas (mō luk'ez) group of islands constituting a province of Indonesia, between Celebes & New Guinea:

c. 32,000 sq. mi.; pop. 790,000; also Molucca Islands

mol. *wt.*, molecular weight

mo-ly' (mō'lē) *n.* [L. < Gr. *mōly*] 1. Classical Myth, an herb of magic powers, as, in Homer's *Odyssey*, that given to Odysseus to protect him from Circe's incantation 2. a European wild garlic (*Allium moly*)

mol-y' (māl'ē) *n.* *clipped form of* **MOLYBDENUM**

mo-lyb-date (mō lib'dāt) *n.* a salt of molybdenic acid

mo-lyb-de-nite (mō lib'dā nīt') *n.* native molybdenum sulfide, MoS₂, a scaly or foliated, lead-gray mineral: the chief ore of molybdenum

mo-lyb-de-num (-nəm) *n.* [ModL. < *molybdēna*, a lead ore, molybdenite, altered < L. *molybdaena*, lead, galena < Gr. *molybdaina* < *molybdos*, lead] a soft, lustrous, silver-white metallic chemical element; used in alloys, windings for electrical resistance furnaces, points for spark plugs, etc.: symbol, Mo; at. wt., 95.94; at. no., 42; sp. gr., 10.2; melt. pt., 2620°C; sublimes at 4507°C

mo-lyb-dic (mō lib'dik) *adj.* *Chem.* designating or of compounds in which molybdenum has a higher valence (usually 3 or 6) than in the corresponding molybdous compounds

mo-lyb-dous (-dās) *adj.* *Chem.* designating or of compounds in which molybdenum has a lower valence than in the corresponding molybdic compounds

mom (mām) *n.* [Colloq.] mother

MOMA Museum of Modern Art

***mom and pop store** (stand, etc.) a small retail business, typically family operated and now often franchised

Mom-ba-sa (mām bā'sā, bas'ā) seaport on the SE coast of Kenya; pop. 180,000

mome (mōm) *n.* [*<* ?] [Archaic] a blockhead; fool

mo-mēnt (mō'mənt) *n.* [ME. < L. *momentum*, movement, impulse, brief space of time, importance < *movimentum* < *movere*, to move] 1. an indefinitely brief period of time; instant 2. a definite point in time or in a series of events 3. a brief time of being important or outstanding 4. importance; consequence [news of great *moment*] 5. *Mech.* a) the tendency to cause rotation about a point or axis b) a measure of this tendency c) the product of a (specified) force, mass, volume, etc. and its perpendicular distance from its axis, fulcrum, or plane 6. *Philos.* any constituent element of a complex entity — **SYN.** see **IMPORTANCE** — *the moment* the present or the immediate future

mo-men-tar-i-ly (mō'mən ter'ē lē) *adv.* 1. for a moment or short time 2. in an instant 3. from moment to moment; at any moment

mo-men-tar-y (mō'mən ter'ē) *adj.* [L. *momentarius*] 1. lasting for only a moment; passing; transitory 2. [Now Rare] recurring every moment; constant 3. likely to occur at any moment — **SYN.** see **TRANSIENT** — *mo-men-tar'i-ness* *n.*

mo-ment-ly (mō'mənt lē) *adv.* 1. from instant to instant; every moment 2. at any moment 3. for a single moment

mo-men-to (mō men'tō) *n.* *erroneous sp. of* **MEMENTO**

***moment of truth** 1. the point in a bullfight when the matador faces the bull for the kill 2. a critical moment or time that tests and reveals one's true self or makes one face the truth

mo-men-tous (mō men'təs) *adj.* of great moment; very important [*a momentous decision*] — *mo-men'tous-ly* *adv.* — *mo-men'tous-ness* *n.*

mo-men-tum (mō men'təm) *n.*, *pl.* -tums, -ta (-tā) [ModL. < L.: see **MOMENT**] 1. the impetus of a moving object 2. strength or force that keeps growing (a campaign that gained *momentum*) 3. *Physics & Mech.* the quantity of motion of a moving object, equal to the product of its mass and its velocity

***mom-ism** (mām'iz'm) *n.* [coined (1942) by Philip Wylie, U.S. writer: cf. **MOM** & **ISM**] excessive devotion to mothers that gives them a domineering role over their children

Momm-sen (mōm'zən; E. mām's'n, -z'n), The-o-dor (tā'dōr) 1817–1903; Ger. historian

mom-my (mām'ē) *n.*, *pl.* -mies *child's term for* **MOTHER**

Mo-mus (mō'mas) [L. < Gr. *Mōmos*, lit., blame, ridicule] Gr. Myth. the god of mockery and censure — *n.* a fault-finder or caviling critic

Mon (mōn) *n.* 1. *pl.* **Mons**, **Mon** any member of a people living in Burma east of Rangoon 2. their Mon-Khmer language

mon (mān) *n.* *Scot. & North Eng. var. of* **MAN**

mon- (mān) same as **MONO-**; used before a vowel

Mon. 1. Monastery 2. Monday 3. Monsignor

mon. 1. monastery 2. monetary

Mo-na (mō'nā) [Ir. *Muadhnaid*; dim. of *muadh*, noble] a feminine name

mon-a-chal (mān'ə k'li) *adj.* [ML(Ec.) *monachalis* < LL(Ec.) *monachus*: see **MONK**] same as **MONASTIC** — *mon'a-chism* (-kiz'm) *n.*

mon-ac-id (mān'as'id) *adj.* *n.* same as **MONOACID**

Mon-a-co (mān'ə kō, mō nā'kō) independent principality on the Mediterranean; enclave in SE France: 1/2 sq. mi.; pop. 23,000

mo-nad (mō'nad, mām'ad) *n.* [LL. *monas* (gen. *monadis*) < Gr. *monas* (gen. *monados*), a unit, unity < *monos*, alone: see **MONO-**] 1. a unit; something simple and indivisible 2. *Biol.* a) any simple, single-celled organism, specif. a simple type of flagellated protozoan or protist b) any of the four nuclei formed at the completion of

submittable

sub-lime (sə blīm') *adj.* [*L. sublimis* < *sub-* up to + *limen*, lintel (hence, orig., up to the lintel): see LIMEN] 1. noble; exalted; majestic 2. inspiring awe or admiration through grandeur, beauty, etc. 3. [Colloq.] outstandingly or supremely such [a man of sublime taste] 4. [Archaic] a) elated; joyful b) proud; lofty; haughty c) upraised: aloft —*vt.* —*limed*, —*lim'ing* [*ME. sublimen* < *MEFr. sublimier* < *ML. sublimare* < *L. sub-* to lift high < the *adj.*] 1. to make sublime 2. to purify (a solid) by heating directly to a gaseous state and condensing the vapor back into solid form —*vi.* to go through this process —*SYN.* see SPLENDID —*sub-lime'ly adv.* —*sub-lime'ness n.*

sub-lim-i-nal (sʌb līm'ə nəl) *adj.* [see SUB- & LIMEN < -AL] below the threshold of consciousness or apprehension; specific: involving or using stimuli that become effective subconsciously by repetition —*sub-lim'i-nal'ly adv.*

sub-lim-i-ty (sə blīm'ə tē) *n.* [*L. sublimitas*] 1. the state or quality of being sublime, majestic, noble, etc. 2. pl. -ties something sublime

sub-lin-gual (sʌb lɪn'gwəl) *adj.* [*ML. sublingualis*: see SUB- & LINGUAL] situated under the tongue

sub-lu-nar-y (sʌb'loo nər'ē) *sub lōon'ər ē*) *adj.* [*ML. sublunaris* < *L. sub-* under + *luna*, the moon] 1. situated beneath the moon; terrestrial 2. earthly; mundane Also *sub-lu'nar*

*sub-ma-chine gun (sʌb mə'shēn'g) a portable, automatic or semiautomatic firearm with a short barrel and a stock, fired from the shoulder or hip

sub-mar-gin-al (sʌb mər'jɪ nəl) *adj.* 1. below minimum requirements or standards (*submarginal housing*) 2. not yielding a satisfactory return; unproductive (*submarginal land*) 3. *Biol.* near the margin of an organ or part —*sub-mar'gin-al-ly adv.*

sub-ma-rine (sʌb mə'ren'ə) *for n. & v. usually sub mən'ren'ə* *adj.* [SUB- + MARINE] being, living, used, or carried on beneath the surface of the water, esp. of the sea —*n.* 1. a submarine plant or animal 2. a kind of warship, armed with torpedoes; guided missiles, etc., that can operate under water —*vt.* —*rin'd*, —*rin'ing* to attack, esp. to torpedo with a submarine

submarine chaser a small, fast naval patrol vessel equipped for use against submarines

sub-ma-rin'er (sʌb mə'ren'ər, sʌb mən'ren'ər) *n.* a member of a submarine crew

*submarine sandwich same as HERO SANDWICH

sub-max-il-la (sʌb mak sɪl'ə) *n.* pl. -lae (ē) -lae [*Mod.L. see SUB- & MAXILLA*] the lower jaw or jawbone

sub-max-il-lar-y (sʌb maks'ələr'ē) *adj.* designating, of, or below the lower jaw; esp., designating or of either of two salivary glands, one on each side, below the inside edge of the lower jaw

sub-me-di-ant (-mē'dē-ənt) *n.* [SUB- + MEDIANT] the sixth tone of a diatonic scale; tone just above the dominant and below the subtonic; superdominant

sub-merge (səb mərj'ē) *vt.* —*merged*, —*mer'ing* [*L. submergere* < *sub-* under + *mergere*, to plunge: see MERGE] 1. to place under or cover with water or the like; plunge into water, inundate, etc. 2. to cover over; suppress; hide 3. to sink below a decent level of life (the submerged people of the slums) —*vi.* to sink or plunge beneath the surface of water, etc. —*sub-mer'gence (-mər'jəns) n.* —*sub-mer'gible (-jə bəl) adj.*

sub-merged (-mərj'ē) *vt.* —*merged*, —*mer'ing* [*< L. submersus*, pp. of *submergere*: same as SUBMERGE] —*sub-mer'sion (-mər'zhan, -shən) n.*

sub-mersed (-mərst'ē) *adj.* submerged; specific: Bot. growing under water

sub-mers-i-ble (səb mər'sə bəl) *adj.* that can be submerged; esp. so as to continue functioning —*n.* any of various ships that can operate under water and are used for exploration, research, etc.

sub-mi-cro-scop-ic (sʌb mi kro skəp'ɪk) *adj.* too small to be seen through a microscope

sub-min-i-a-ture (sʌb mɪn'ə tʃər) *adj.* designating or of a very small camera, electronic component, etc., smaller than one described as "miniature"

sub-min-i-a-tur-ize (-iz'ē) *vt.* —*ized*, —*iz'ing* to construct (something) on a subminiature scale —*sub-min'i-a-tur'iz-a-tion n.*

sub-miss (-mɪs'ē) *adj.* [Archaic] submissive; humble

sub-mis-sion (-mɪsh'ən) *n.* [*ME. < OFr. < L. submissio* < *sub-*missive, pp. of *submittere*] 1. the act of submitting; yielding; or surrendering 2. the quality or condition of being submissive; resignation; obedience; meekness 3. the act of submitting something to another for decision, consideration, etc. 4. *Law* an agreement whereby parties to a dispute submit the matter to arbitration and agree to be bound by the decision

sub-mis-sive (-mɪs'iv) *adj.* [*< L. submissus*, pp. of *submittere* (see fi.) + -IVE] having or showing a tendency to submit without resistance; docile; yielding; —*sub-mis'sive-ly adv.* —*sub-mis'sive-ness n.*

sub-mit (-mɪt'ē) *vt.* —*mit'ted*, —*mit'ting* [*ME. submitten* < *L. submittere* < *sub-* under, down + *mittere*, to send: see MISSION] 1. to present or refer to others for decision, consideration, etc. 2. to yield to the action, control, power, etc. of another or others; also, to subject or allow to be subjected to treatment, analysis, etc. of some sort: often used reflexively 3. to offer as an opinion;

suggest; propose.—*vt.* 1. *a)* to *yi* etc. of another or others; give in subjection (*to* treatment, analysis another's judgment or decision 3 ent; humble, etc.—*SYN.*: see *SUBJ*)
sub-mit-tal *n.*—**sub-mit-t' adj.**—**sub-mon-tane** (sub/mān'tān) *adj* see *MOUNTAIN* 1. located at the mountain range 2. of or character
sub-mul-ti-ple (sub mul'ti p'l) 1. number that will divide another's divisor (of a specified number) 2. **sub-nor-mal** (n-ōr'm'l) *adj.* below normal, esp. in intelligence —**sub-nor-mal'ty** *f.* (mal'f tē) *n.*
sub-nu-cle-ar (n-ōō'klē ar, -nōf of any of the particles within the sub-o-ce-an-ic (sub'ō shē an'ik) on or beneath the ocean-floor
sub-or-bit-al (sub'ōr'bit'l) *adj.* spacecraft in which the spacecraft range trajectory instead of going the orbit of the eye
sub-or-der (sub'ōr'dar) *n.* any n order of plants or animals —**sub-or-di-nate** (sə bōr'dā-nit; for *ML*, *subordinate*, pp. of *subordinare*, to order: see *ORDAIN*) below another in rank, power, im 2. under the power or authority or submissive 4. *Gram.* function or adverb within a sentence (A *SUBORDINATE CLAUSE* —*n.* a sub-
—*ut.* -na'ted, -na'ting 1. to position; treat as less important or obedient or subservient (*to*); cor di-nate-ly *adv.* —**sub-or-di-na-ti subordinate clause** in a complex cannot function syntactically as itself; dependent clause: distinguish (Ex.: She will visit us *if she can*)
subordinating conjunction *a* subordinate words, phrases, or sentence element (Ex.: *if, as, so*, also *subordinate conjunction*)
sub-or-di-na-tion (sə bōr'dā nā'sh-n) *n.* being subordinated. 2. (N submission to rank, power, or aut
sub-orn (sə bōr'n) *v.* [*MF*, *sub* furnish or supply, instigate, incite + *ornare*, to furnish, adorn: see bring about through bribery or to induce or instigate (another) *esp.* to commit perjury —**sub-orn sub-or-na-tion** (sub'ōr nā'shān) being suborned; *esp.* the crime commit perjury (subornation of
sub-ox-ide (sub'āk'sid) *n.* an oxid small proportion of oxygen
sub-phy-lum (sub'fē'lēm) *n.* *pl.* subdivision of a phylum
sub-plot (n-plot) *n.* a secondary p
sub-po-e-na (sə pē'nā) *n.* (*ML*, *sub poena*, lit., under penalty: see legal order directing a person to testimony, show specified records, 1. to summon with such an order fined records, documents, etc.)
Also *sp.* **sub-pe-na**
sub-pop-u-la-tion (sub'pāp yā lā'sh) a population, with common, dist
sub-prin-ci-pal (sub prin'se p'l) principal in a school, etc. 2. a 3. *Mus.* an open diaphan subbas
sub-pro-fes-sion-al (sub'prō fē PARAPROFESSIONAL
sub-re-gion (sub'rē'jān) *n.* any of *esp.* with reference to plant and a
sub-rep-ri-tion (sə rep'shān) *n.* [*l* pp. of *subripere*, *surreipere*, to surreptitious] 1. the fraudulent representation of facts so as to clestial dispensation 2. a false sub-rogation —**sub-rep-ti-tion**
sub-ro-gate (sub'rə gāt') *vt.* *g* subrogatus, *subrogatus*: see *SURRO person* for another
sub-ro-ga-tion (sub'rə gā'shān) *ML*, *subrogatio* < *L*, *subrogatus* substitution of one creditor for transference of the claims and rig
sub-ro-sa (sə rō'sā) [*L*, lit., an symbol of secrecy] secretly; priva
sub-rou-tine (sub'rōō tēn') *n.* 1. tions. often used repeatedly, that

fat, âpe, căr; ten, even; is, bite; gô,
e for a in ago, e in agent, i in sanity, c
tă, Fr. duc; r, Fr. cri; H, G. ich; k

apparent to the investigator as he gains facility using the diagrams.

Denoting configurational average by a subscripted angular bracket, we have

$$\langle\langle E^2 \rangle\rangle_c = N^{-1} \sum_{i=1}^N \langle H_{ii}^2 \rangle_c + 2 \langle H_{ii} H_{i,i+1}^2 \rangle_c + 2 \langle H_{ii} H_{i,i-1}^2 \rangle_c + \langle H_{i+1,i+1} H_{i,i+1}^2 \rangle_c + \langle H_{i-1,i-1} H_{i,i-1}^2 \rangle_c \quad (5A)$$

We may eliminate the summation and the factor N^{-1} since terms whose indices can be made identical by replacing i by $i+j$ are equal to each other. Strictly speaking, this means that we are neglecting end effects

and are considering an infinite chain of square wells. With this assumption, we may write

$$\langle\langle E^2 \rangle\rangle_c = \langle H_{ii}^2 \rangle_c + 2 \langle H_{ii} H_{i,i+1}^2 \rangle_c + 2 \langle H_{ii} H_{i,i-1}^2 \rangle_c + \langle H_{i+1,i+1} H_{i,i+1}^2 \rangle_c + \langle H_{i-1,i-1} H_{i,i-1}^2 \rangle_c \quad (6A)$$

By the same argument we could combine, for instance, terms two and five, although this simplification is not used in Eq. (27). However, terms two and three are not identical, nor is

$$\langle H_{ii} H_{i,i+1}^2 \rangle_c = \langle (H_{ii}) \rangle_c \langle (H_{i,i+1}^2) \rangle_c$$

These latter points can be proved by direct computation using the probability distribution functions (25).

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Solubility of Carbon in Silicon and Germanium

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The solubility of carbon in silicon has been measured over the temperature range 1560 to 2900°C. The enthalpy of solution is 59 ± 3 kcal/mole. A phase diagram for the system Si—C is presented, embodying these solubility data as well as the results of other high-temperature experiments with silicon carbide. It is found that SiC possesses a peritectic point at $2830 \pm 40^\circ\text{C}$. These studies were carried out in argon at pressures as high as 35 atmos. Solubilities of carbon in germanium were measured in the temperature range 2780 to 3170°C, at argon pressures up to 55 atmos, and a tentative phase diagram is given.

EXPERIMENTS WITH SILICON

Introduction

NEITHER the solubility of carbon in silicon at high temperatures, nor the phase diagram of the system Si—C is known. Nowotny *et al.*,¹ in working with the ternary system Mo—Si—C, proposed two possible phase diagrams for the system Si—C. These were for pressures less than one atmosphere and were based on a limited number of experimental points so that a decision between the two could not be made. The solubility of C in liquid Si has been measured by Hall² up to 1710°C. Others³ have found Si as an impurity in natural diamond, although the conditions of formation are of course unknown. We present here some solubility measurements in liquid Si up to 2900°C, and a proposed phase diagram for the system.

Experimental Procedure

The experiments were carried out in a 50-kw graphite resistance furnace heated by 20 v maximum, 60-cy

ac. This was enclosed in a water-cooled pressure vessel. Argon at a pressure of 35 atmos was used to reduce the tendency of the silicon to evaporate. The vapor pressure of silicon is about one atmosphere at 2800°C over both Si⁴ and SiC.⁵ Figure 1 shows the arrangement of the silicon container, heater, and shields which were used above 2000°C. The entire assembly was of graphite and was supported from the current terminals. On the basis of melting point measurements on iridium wires, the temperature variation over the container was believed to be not over $\pm 25^\circ\text{C}$ from the value at the point of measurement. The mounting of the container within the heater tube was such that no current passed through the Si charge. In this way, changes in the state of the Si, and hence in its electrical resistivity, did not affect the temperature distribution.

The Si was du Pont hyperpure grade, ground with a Pyrex mortar and pestle, screened, and leached with HF to remove any glass contamination in the Si from the mortar. The graphite container was filled with 100 mesh Si (about 0.5 g) and closed with a graphite end plug. No carbon was added; the carbon which was

¹ Nowotny, Parthe, Kieffer, and Benesovsky, *Monatsh. Chem.* 85, 255 (1954).

² R. N. Hall, *J. Appl. Phys.* 29, 914 (1958).

³ F. G. Chesley, *Am. Mineralogist* 27, 20 (1942); F. A. Raal, *ibid.* 42, 354 (1957).

⁴ R. E. Honig, *J. Chem. Phys.* 22, 1610 (1954).

⁵ Drowart, de Maria, and Inghram, *J. Chem. Phys.* 29, 1015 (1958).

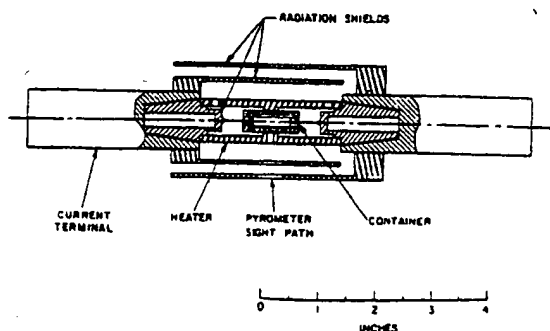


FIG. 1. Graphite resistance furnace (axial cross section).

taken into solution by the liquid silicon came from the walls of the container.

After assembly of the furnace, the pressure vessel was evacuated; it was then refilled twice with argon to a pressure of 20 atmos, vented to atmospheric pressure each time, and finally filled to the operating pressure. A manostat maintained the pressure within ± 1 atmos.

The length of time that the charge was kept at the operating temperature was a compromise. A long time ensured equilibrium, but for long times the Si loss became excessive with graphite containers. Usually temperature was kept constant at the desired value for periods of one-half to two minutes. No dependence of the C solubility upon the duration of run was observed. The charge was cooled as rapidly as possible after the run by simply shutting off the power to the furnace. Initial cooling rates of $50^\circ\text{C}/\text{sec}$ were typical.

Analysis

The carbon which was in solution in the liquid silicon at high temperatures crystallizes as SiC upon cooling. The Si slug containing the SiC was recovered by oxidizing away the graphite reaction vessel at 1100°C in an O_2 atmosphere. Less than 0.2 mg of SiO_2 was produced by this treatment. The original charge of Si could be accounted for in four portions:

1. Some had reacted with the container, forming a microcrystalline mass of SiC within the pores of the graphite container. Use of a dense graphite ($1.90 \text{ g}/\text{cm}^3$)* for the containers was necessary to avoid excessive losses in this way. This loss occurred mainly during the furnace warmup after the silicon liquified, and in unfavorable cases amounted to over 50 percent of the initial charge.

2. Some remained within the container as elemental Si, frozen into a small slug. This generally ranged from 10 percent to 60 percent of the initial charge of 500 mg, depending on the porosity of the container.

Some precipitated as SiC during cooling of the furnace. Most of the SiC was enclosed within the frozen

mass of Si, although some may have segregated to the surface of the Si. No free C remained within the piece of silicon, as determined both by microscopic examination and by chemical analysis of the residue which remained after the silicon was removed by etching.

4. Little if any Si was lost by evaporation because of the blanketing effect of the argon and the low porosity of the graphite container.

The slug of elemental Si containing the precipitated silicon carbide was lightly sandblasted to remove SiC which was formed in the walls of the graphite container and adhered to the surface of the slug. A thin layer of silicon (less than 25μ) was removed from the slug during the sandblasting. After the sandblasted slug was weighed, the Si was etched away in a mixture of 1 part HF to 3 parts HNO_3 . The SiC residue, which was not attacked by the etch, was dried and weighed. The carbon solubility at the operating temperature of the furnace was computed from the ratio of the mass of SiC to the mass of Si in the slug.

The major source of error in this procedure was the separation of the SiC formed by surface reaction with the container from that formed by reaction with dissolved carbon. It is felt that the experiments done in graphite gave a lower limit to the solubility of C in Si, because some dissolved SiC which was segregated to the surface during cooling may have been lost in sandblasting. Because only a thin layer was removed it is believed that at least 90 percent of the SiC is retained in the slug. Also, from the reproducibility of experiments one concludes that this loss was not serious.

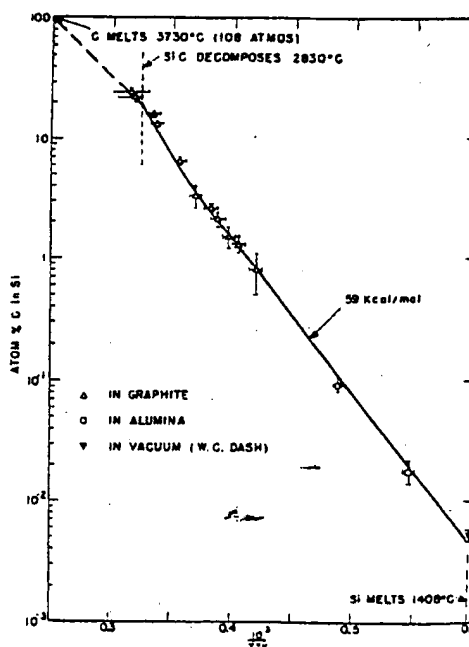


FIG. 2. The solubility of C in Si as a function of temperature.

Two of the measurements at 2000°C were made in different heaters and this change was noticeable (10–20 g) was needed of carbon. In the latter case SiC was included in order to determine the content of the Si melt. SiC which was dissolved. The total amount of SiC was 5 mg. These experiments were in order to provide a check on the data in graphite. These data were used because all the SiC was considered to have gone to redeposition of SiC or had occurred during the measurements performed in graphite is. A measurement of the SiC was made by W. C. Dash using a pedestal clamped at the bottom of a portion melted. The SiC of about 4.1 mg was melted and sank until it reached the bottom. After about 10 minutes the SiC which surrounded the etching and again the weight loss and the mass solubility to be found. silicon at the melting point.

Temperature

All temperature measurements were made in Leeds & Northrup type thermocouples which had been recently calibrated. Errors are known to within 1 percent on the temperature (L) scale. Corresponding scales have at most a 1 percent transmission through the furnace. These measurements were made separately inside the furnace as a check on the measurements made in place and removed. No correction to be made in the operation of the surface of the furnace at the bottom of a hole is deep. The rough walls of the furnace and the solubility of graphite both tend to make the furnace wall of the Si container a blackbody. The temperature of the container was determined by direct measurement of the container which had a hole. This hole was so positioned that it was not possible to see the bottom of the hole.

* We are indebted to R. L. Shepard, of the National Carbon Company Research Laboratories, for some samples of a suitable high-purity graphite.

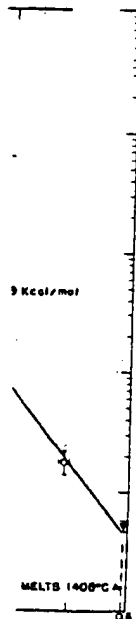
* W. C. Dash, J. Appl. Phys.

have segregated to the end within the piece of microscopic examination residue which remained etching.

vapor because of and the low porosity

ning the precipitated asted to remove SiC he graphite container slug. A thin layer of oved from the slug he sandblasted slug ay in a mixture of residue, which was d and weighed. The temperature of the atio of the mass of

s procedure was the urface reaction with reaction with dis- xperiments done in olubility of C in Si, as segregated to the een lost in sand- was removed it is the SiC is retained ucibility of experi- as not serious.



on of temperature.

Two of the measurements at temperatures below 2000°C were made in the same furnace, but with a different heater and an Al_2O_3 container (see Fig. 2). This change was necessary because a larger melt (10–20 g) was needed to dissolve a measurable amount of carbon. In the latter experiments, a weighed crystal of SiC was included in the melt, and its weight loss was used to determine the solubility. As a check, the SiC content of the Si melt was also found. About 2/3 of the SiC which was dissolved from the crystal was recovered. The total amount of SiC lost from the crystal was 5–10 mg. These experiments in ceramic containers were done in order to provide a comparison with those carried out in graphite. These data are upper limits to the solubility, because all the SiC lost from the weighed piece was considered to have gone into solution even though some redeposition of SiC on the walls of the container may have occurred during the run. The agreement between the measurements performed in Al_2O_3 and those performed in graphite is good, as can be seen from Fig. 2. A measurement of the solubility at the Si melting point was made by W. C. Dash. This was performed in vacuum using a pedestal technique.⁶ A vertical bar of Si, clamped at the bottom, was heated at the top until a portion melted. Then a carefully weighed crystal of SiC of about 4.1 mg was inserted from above into the melt and sank until it rested at the liquid-solid interface. After about 10 min the melt was refrozen. The Si which surrounded the SiC crystal was removed by etching and again the SiC crystal was weighed. The weight loss and the mass of Si in the melt enabled the solubility to be found. The carbon solubility in liquid silicon at the melting point is $3.0 \pm 0.3 \times 10^{-18}$ A/cm³.

Temperature Measurement

All temperature measurements were made with a Leeds & Northrup type 8622-C optical pyrometer which had been recently standardized. Calibration errors are known to within $\pm 2^\circ\text{C}$ on the low-temperature (*L*) scale. Correspondingly the higher temperature scales have at most a $\pm 10^\circ\text{C}$ error. Measurements of transmission through the sight tube window of the furnace were made separately using a tungsten strip lamp inside the furnace as a source, with the window alternately in place and removed. This allowed an accurate correction to be made for transmission losses. During operation the surface of the Si container was visible at the bottom of a hole in the heater tube two diameters deep. The rough walls of the hole and the high emissivity of graphite both tended to make the hole equivalent to a blackbody. The temperature drop across the wall of the Si container was found as a function of temperature by direct measurement on an empty container which had a hole through to the interior. This hole was so positioned that both internal and

⁶ W. C. Dash, J. Appl. Phys. 29, 736 (1958).

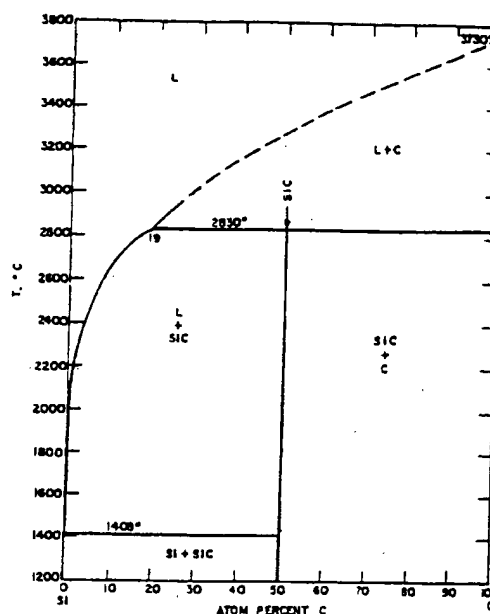


FIG. 3. Phase diagram of the system Si-C for pressures above 108 atmos.

surface temperatures could be measured at the same time. A useful check on the pyrometer was obtained from a plot of heater current vs absolute temperature squared. Above about 1700°C this plot was linear, indicating that radiation was by far the most important mechanism of energy loss.

As a final test, the melting points of Ir (2454°C) and Mo (2622°C) were measured, care being taken to avoid contact between the metal and carbon. We obtained 2446°C and 2634°C, respectively.

Pyrometry errors may be estimated as follows: setting and calibration errors on the instrument, $\pm 10^\circ\text{C}$; window transmission uncertainty, $\pm 20^\circ\text{C}$; and error in the temperature drop across the reaction container wall, $\pm 10^\circ\text{C}$.

Results

The solubility of C in Si as a function of temperature is given in Fig. 2. Some curvature may be noted at concentrations of five atom percent and more, presumably because the solution is nonideal. We find the solubility to be about 1/5 of that reported by Hall.² Recent experiments by J. H. Racette⁷ of this laboratory, in a fused quartz apparatus similar to that used by Hall, indicate that this discrepancy was most likely due to the presence of sufficient oxygen in the melt in Hall's experiment to remove much of the dissolved carbon as CO or CO_2 . The oxygen presumably came from the quartz (SiO_2). In that case the carbon solubility would have appeared to be too high because the SiC crystal placed in the melt continuously lost carbon. Racette's

⁷ J. H. Racette (private communication).

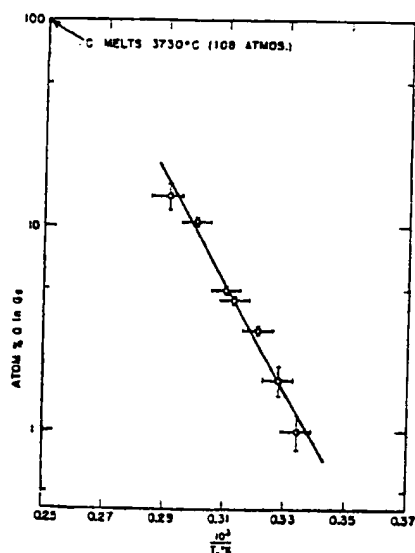


FIG. 4. The solubility of C in Ge as a function of temperature.

experiments show an apparent increase in solubility as the duration of the experiment is increased.

The linear portion of the curve below 1A% carbon as a slope corresponding to an enthalpy of solution of 9 kcal/mole, as calculated from the expression

$$x = \exp\{(\Delta H/R)[(1/T) - (1/T_m)]\}. \quad (1)$$

In this equation, x is the fractional atomic concentration of carbon in silicon at temperature T , ΔH is the enthalpy of solution, R is the gas constant per mole, and T_m is the temperature intercept at unit carbon fraction obtained by extrapolation of the linear portion of the solubility curve. Depending on how the line is drawn through the data points, ΔH may vary ± 3 kcal/mole.

In the course of these experiments it seemed worthwhile to explore as much of the phase diagram of the system Si-C as was accessible. Attempts were made to melt SiC in graphite containers similar to those used for the solubility experiments. The ambient pressure was well in excess of the decomposition pressure in all experiments. A transformation of some originally light green, type 6H, hexagonal, 120-mesh, SiC grain to a markedly different structure occurred at temperatures of $2830 \pm 40^\circ\text{C}$ and above. The SiC found after the transformation was cubic, as analyzed by x-rays. Heating to lower temperatures resulted only in a sintering together of the grains without any gross structural change. This experiment was repeated with cubic SiC grain as the initial charge. It decomposed at the same temperature, and the SiC found after the transformation was also cubic. From this, and from evidence gained from other experiments with SiC at elevated temperatures, we conclude that SiC decomposes without congruent melting, resulting in a silicon-rich liquid in

equilibrium with graphite at temperatures from 2830°C to at least 3160°C . The liquid composition at this latter temperature is approximately 50 atom percent carbon. When the liquid resulting from the decomposition of SiC is cooled below the decomposition temperature, the Si reacts with the dissolved and suspended carbon to form the cubic SiC found by x-ray analysis.

Experiments have also been performed in which cubic SiC grains have been heated in graphite tubes in this furnace to temperatures slightly below 2830°C . At the end of a run lasting several minutes the original cubic SiC is still present although slightly sintered together. Other experiments have been conducted in this apparatus in which both cubic and hexagonal crystals of SiC have been grown side by side from the vapor phase at temperatures of 2600°C . These growing experiments lasted several hours. It is therefore concluded that cubic SiC is stable at these temperatures and does not readily transform into hexagonal SiC in contrast to the conclusions of Baumann.⁹ However, it should be noted that the conditions in the present experiments are not the same as those found in commercial SiC furnaces.

Difficulty was encountered in attempting to extend the solubility measurements above 3000°C . Even the high-density graphite containers would not hold liquid silicon at these temperatures. Furthermore the interpretation of the chemical analysis became difficult because the sample after cooling to room temperature usually contained silicon carbide, silicon, and carbon in a carbon container.

The phase diagram shown in Fig. 3 incorporates the

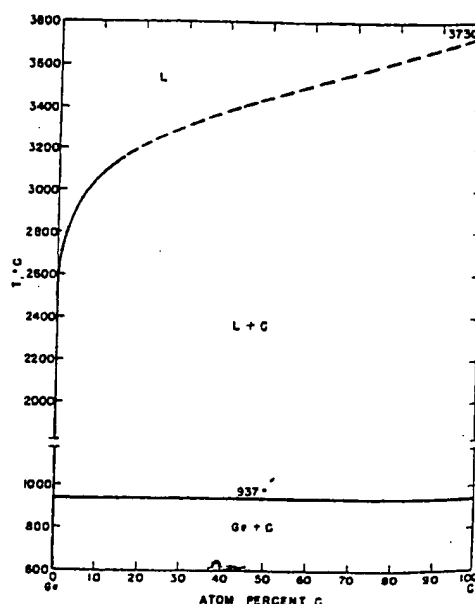


FIG. 5. Tentative phase diagram of the system Ge-C for pressures above 108 atmos.

⁹ H. N. Baumann, J. Electrochem. Soc. 99, 109 (1952).

findings described given by Basset,⁹ as by Pell.¹⁰ It confirms grams proposed by sen.¹¹ Note that it assumed to be high are always present. pure carbon at 3730°C over carbon at the the required pressure: 15 atmos a temperature reached.⁴

EXPERIMENT

The solubility of apparently not been previously shown in Fig. containers to temperatures and have analyzed the limit of 3170°C was germanium evaporation maximum argon pressure below 2780°C the available limited volume of liquid measure accurately. refined; the container scopic graphite with a The samples were graphite containers. to the graphite, and After weighing, the sample part HF in three parts precipitated graphite

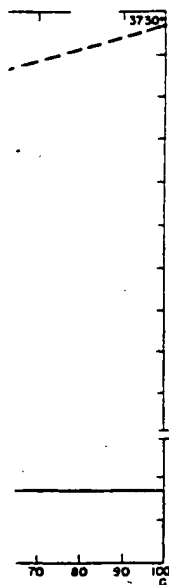
⁹ J. Basset, J. phys. radi
¹⁰ E. M. Pell, J. Phys. C
¹¹ M. Hansen, Constitution, p. 378.

temperatures from 2830°C in position at this latter atom percent carbon. the decomposition of position temperature, and suspended carbon ray analysis.

formed in which cubic graphite tubes in this below 2830°C. At the test the original cubic tubes sintered together. conducted in this apparatus. gonal crystals of SiC in the vapor phase at growing experiments concluded that cubic ; and does not readily contrast to the con- should be noted that eriments are not the SiC furnaces.

attempting to extend e 3000°C. Even the ould not hold liquid hermore the interpre- ame difficult because temperature usually id carbon in a carbon

g. 3 incorporates the



system Ge-C for pres-

99, 109 (1952).

findings described above, the melting point of carbon given by Basset,⁹ and the melting point of silicon given by Pell.¹⁰ It confirms the general features of the diagrams proposed by Nowotny *et al.*¹ and quoted by Hansen.¹¹ Note that in Fig. 3 the ambient pressure is assumed to be high enough that solid or liquid phases are always present. This will be at least 108 atmos for pure carbon at 3730°C, since this is the vapor pressure over carbon at the triple point.⁹ At lower temperatures the required pressure will be less. With a pressure of 35 atmos a temperature of about 3400°C can be reached.⁴

EXPERIMENTS WITH GERMANIUM

The solubility of carbon in germanium has apparently not been previously reported. Using the furnace shown in Fig. 1, we have heated Ge in graphite containers to temperatures between 2780°C and 3170°C and have analyzed the results. The upper temperature limit of 3170°C was set by the requirement that the germanium evaporation rate be kept low with a maximum argon pressure of 55 atmos. At temperatures below 2780°C the amount of carbon dissolved in the limited volume of liquid germanium was too small to measure accurately. The Ge had been previously zone refined; the containers were machined from spectroscopic graphite with a density of 1.7 g/cm³.

The samples were recovered by cracking apart the graphite containers. The germanium did not adhere to the graphite, and was in the form of a shiny slug. After weighing, the slug was etched in a mixture of one part HF in three parts HNO₃. The residue was partly precipitated graphite in the form of small flakes and

partly Ge. Apparently the etch was unable to leach all the Ge from the graphite flakes because of the hydrophobic nature of graphite. The residue was therefore roasted in Cl₂ at 1000°C in a quartz crucible to remove the Ge by the formation of volatile GeCl₄. After the remainder of the residue was weighed, it was then burned in O₂ at 1000°C to form CO₂. No evidence of GeO or GeO₂ was found after the oxidation, indicating that the residual Ge had indeed been removed from the graphite.

The results are shown in Fig. 4. If the solubility line were extrapolated to the melting point of Ge, the concentration of C in the liquid would be about 10⁸ atoms per cm³. This is probably a lower limit. The carbon-germanium system is nonideal and such an extrapolation is inaccurate. This extrapolated solubility is a factor of about 10¹⁰ less than the carbon concentration in liquid silicon at its melting point. No evidence of compound formation between carbon and germanium was found up to temperatures of 3170°C. The C-Ge phase diagram therefore appears to be quite simple, and a tentative sketch is shown in Fig. 5. A simple eutectic point probably exists very slightly below the melting point of Ge at 937°C.¹² This is qualitatively similar to the C-Pb and C-Sn systems as deduced from the solubility studies of C in Pb and in Sn at temperatures up to 2300°C.¹³

ACKNOWLEDGMENTS

We are much indebted to W. C. Dash for his measurement of the solubility of C in Si at the Si melting point. Also, we wish to thank R. N. Hall and J. H. Racette for many helpful discussions.

⁹ J. Basset, *J. phys. radium* 10, 217 (1939).

¹⁰ E. M. Pell, *J. Phys. Chem. Solids* 3, 77 (1957).

¹¹ M. Hansen, *Constitution of Binary Alloys* (1958), second edition, p. 378.

¹² E. S. Greiner and P. Breidt, *Trans. Am. Inst. Mining Met. Engrs.* 203, 187 (1955).

¹³ O. Ruff and B. Bergdahl, *Z. anorg. u. allgem. Chem.* 106, 76 (1919).

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THE Si-C AND Ge-C PHASE DIAGRAMS*

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Abstract—The solubility of carbon in silicon has been measured over the temperature range 1408°C to 2900°C. The enthalpy of solution is 59 ± 3 kcal/mol. A phase diagram for the system Si-C is presented, embodying these solubility data as well as the results of other high temperature experiments with silicon carbide. It is found that SiC possesses a peritectic point at $2830 \pm 40^\circ\text{C}$. These studies were carried out in argon at pressures as high as 35 atm. Solubilities of carbon in germanium were measured in the temperature range 2780°C to 3170°C, at argon pressures up to 55 atm.

IN the literature on silicon carbide, one finds reference to the fact that SiC decomposes at high temperatures. The temperature at which this occurs has been variously cited, and values from 2200 °C^{4,5} to 2700 °C^{8,9} may be found. This wide range, and the fact that most, if not all of these studies were done under non-equilibrium conditions, leads to the belief that a true decomposition of bulk SiC is not what has been observed, but instead a dissociation of the surface caused by preferential evaporation of silicon from the SiC. The heat of formation data of Humphrey *et al.*⁷, when extrapolated to higher temperatures, indicate that decomposition may occur near 2600 °C. If SiC could be melted without the occurrence of bulk decomposition, then crystals could be grown from the melt. Even if SiC does not possess a melting point, further knowledge of the phase diagram of the system Si-C would be useful in crystal growing work.

To investigate the behavior of SiC at elevated temperatures and pressures, the furnace shown in Fig. 1 was built. A water-cooled pressure vessel surrounds the graphite furnace assembly seen in the center. The vessel will hold a pressure of as much as 1000 lb/in.²; this dense atmosphere retards the evaporation of silicon which would otherwise be quite troublesome.

Figure 2 shows a cross-section of the furnace assembly. All the parts are made of graphite. The two current terminals support a tubular heater, two radiation shields, and a container which was used in the experiments on the solubility of carbon in silicon. The container is supported in such a way that it carries no current. About 50 kW is available to heat this assembly, enabling temperatures of about 3500 °C to be reached.

Optical pyrometry is the only reasonably accurate way of measuring temperature above 1900 °C, which is the limit for platinum-alloy thermocouples. A sight tube and window are provided in the side of the pressure vessel, and openings are left in the radiation shields and heater tube to allow measurement of the internal temperature of the furnace. Corrections to the pyrometer reading must be made for window transmission

* A more complete account of this work has been accepted for publication in the *Journal of Chemical Physics*.

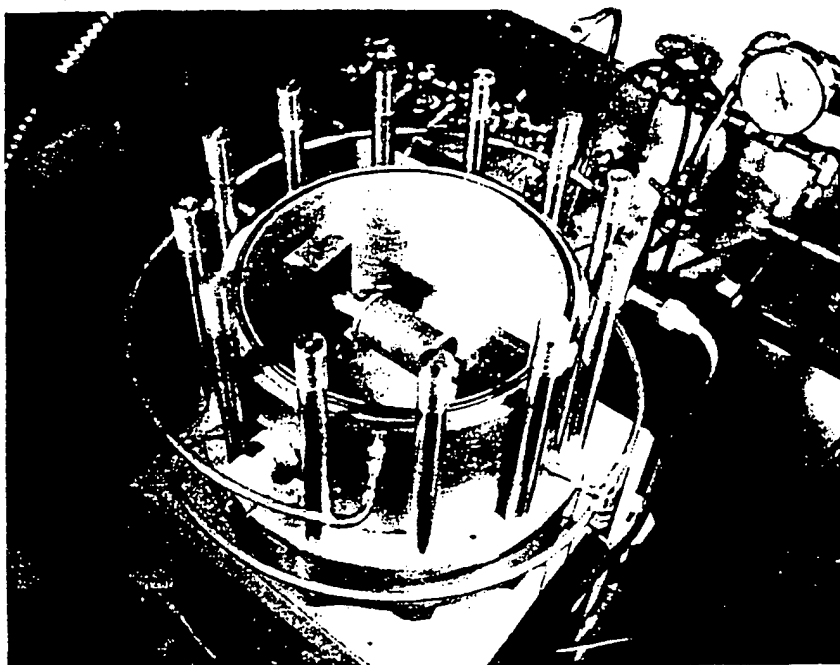


FIG. 1. Interior view of pressure vessel, showing high-temperature furnace in place.

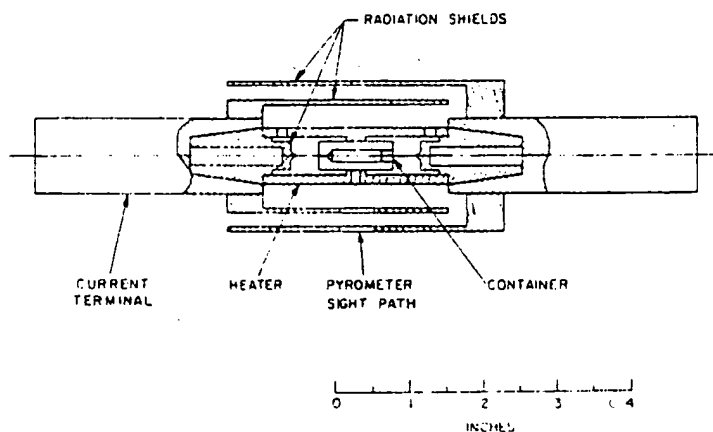


FIG. 2. Cross-section sketch of furnace assembly.

and the emissive properties of the surface whose temperature is to be found. As a check, the melting points of Ir and Mo were measured. The temperatures found were within $\pm 10^\circ\text{C}$ of the published values. Under ordinary circumstances, temperature measurements are believed to be within $\pm 30^\circ\text{C}$ of the true value except at the very highest temperatures (say 3000° or more) where errors may amount to 50°C .

To determine the solubility of C in Si, the inner container was filled with Si. The container was sealed with a tight fitting plug, assembled into the furnace and heated to some temperature between 2100 °C to 2900 °C. Thermal equilibrium was achieved quite rapidly, and the sample was held at a fixed temperature for one or two minutes, after which the power was shut off. Then the furnace temperature decreased rapidly by radiation cooling and the Si inside froze. The graphite container was removed from the furnace and burned away from the Si by heating in O_2 . The original charge of Si can be accounted for as follows:

1. Some had reacted with the container, forming a micro-crystalline mass of SiC within the pores of the graphite.

2. Some remained within the container as elemental Si, frozen into a slug. This ranged from 10 to 60 per cent of the initial charge of 500 mg.

3. Some combined with the dissolved C in the liquid Si, and precipitated within the frozen slug of Si as SiC. No free C was found inside the slug of Si.

4. Very little, if any, Si was lost by evaporation because of the blanketing effect of the high pressure argon atmosphere, and the low permeability of the high density graphite.

The loosely adhering SiC, formed by reaction with the container, was carefully removed from the frozen slug. The slug was then weighed, etched in HF-HNO₃ to remove the Si, and the remaining SiC found inside the Si slug was weighed. With these data the solubility could be found for the temperature in question.

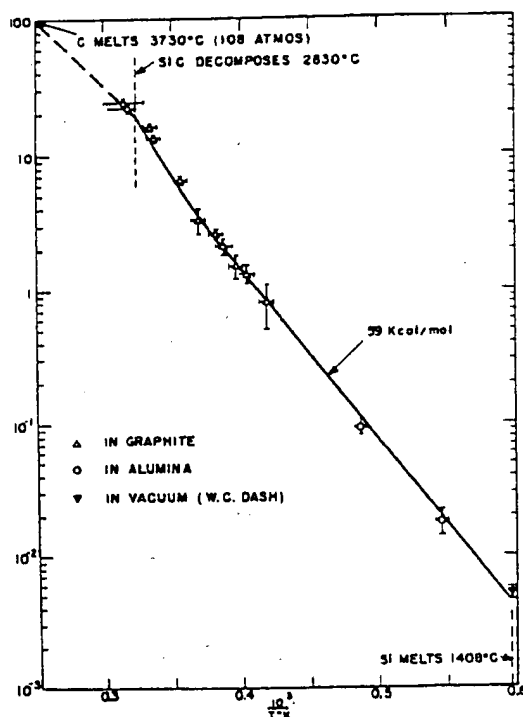


FIG. 3. The solubility of carbon in silicon as a function of reciprocal temperature.

Below 2000 °C the solubility is so low that an accurate determination with 500 mg of Si solvent was impossible. Two runs of 10 and 20 g melts were done in recrystallized alumina crucibles, with a different furnace arrangement in the pressure vessel. Here a weighed single crystal of SiC was included in the melt, and its weight loss combined with the known weight of Si in the melt enabled the solubility to be found.

Finally, Dash² performed a measurement of the solubility at the Si melting point, again by measuring the weight loss of a single crystal of SiC. His melt of Si was supported on top of a pedestal¹ of Si in vacuum. The SiC crystal rested on the liquid-solid interface, so there is little question of temperature measurement accuracy.

These data are all summarized in Fig. 3. The consistency of the results of the various experiments is good. Two points of particular interest are the solubilities of carbon in liquid Si at the Si melting point, 5×10^{-3} atom per cent (5×10^{17} C/cm³ Si), and at the decomposition temperature of 2830 °C. This is the maximum temperature at which SiC could be grown from a Si solution, and the carbon content of the melt here is 19 atom per cent. This high solubility indicates that growth from solution is feasible. The vapor pressure of Si over Si⁶ and over SiC³ at this temperature is of the order of 1 atm and rising rapidly, so the need for a pressurized atmosphere to maintain stable conditions for any length of time is apparent.

Another way of plotting these data is on a temperature vs. composition phase diagram, Fig. 4. The behavior of SiC at the decomposition temperature is more clearly seen. This temperature, 2830 °C, was found by heating samples of SiC grain in containers such as

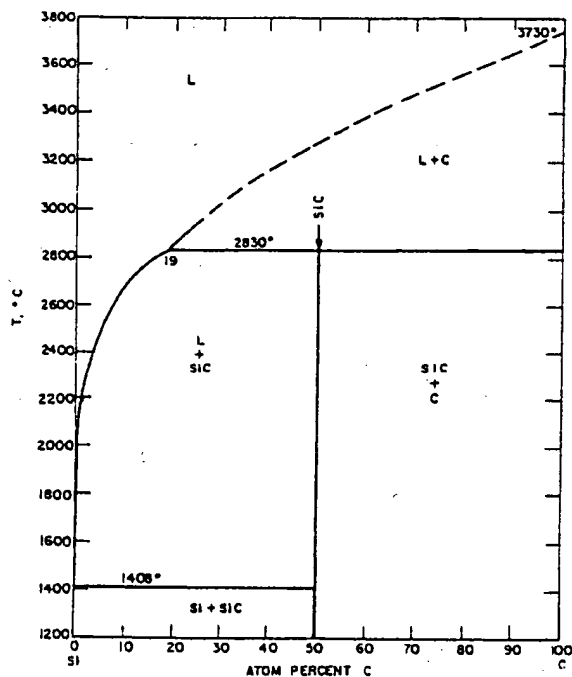


FIG. 4. Phase diagram of the binary system Si-C.

were used for the solubility measurements. All samples whose temperature exceeded this value were decomposed, while none were affected which were cooler. Two fortunate trials, one with hexagonal SiC and one with cubic, were half decomposed and half not, so the temperature was determined as closely as pyrometry errors will allow.

Some measurements have also been performed on the C solubility in Ge, in a manner similar to that used for Si. Figure 5 shows a tentative Ge-C phase diagram based on these measurements. Solubilities of 12 per cent at 3170 °C were reached, where the Ge vapor pressure is several atmospheres. No evidence of compound formation was seen. A very rough extrapolation of these data to the melting point of Ge can be made, indicating that the C solubility in liquid Ge at this temperature is of the order of 10^{10} C/cm³ Ge.

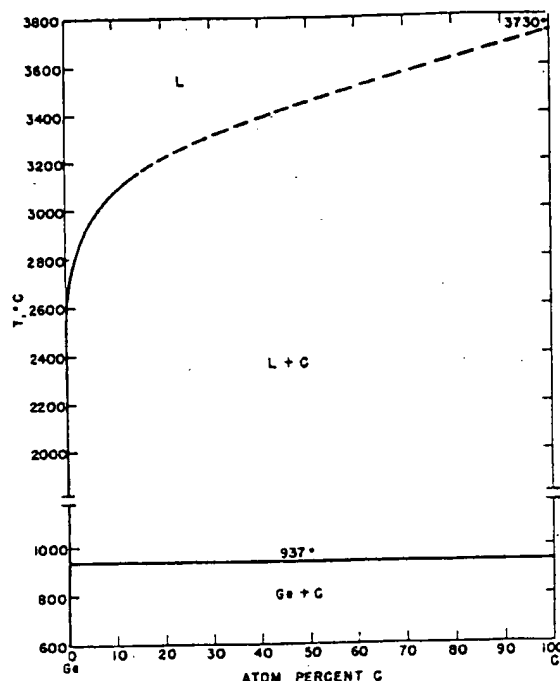
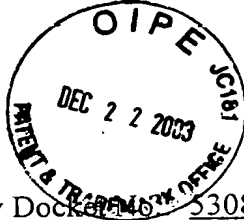


FIG. 5. Tentative phase diagram of the binary system Ge-C.

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- ⁸ O. RUFF and M. KONSCHAK, **32**, 515-525 (1926).
- ⁹ F. J. TONE, *Ind. Eng. Chem.* **23**, 1312-1316 (1931).



Attorney Docket No. 5308-159

PATENT

...IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re: Thomas G. Coleman

Serial No.: 09/931,537

Filed: August 16, 2001

For: SILICON CARBIDE SUBLIMATION SYSTEMS
AND ASSOCIATED METHODS

Group: 1765

Examiner: M. Song

Confirmation No. 3269

December 17, 2003

COPY

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Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

APPELLANTS' BRIEF ON APPEAL UNDER 37 C.F.R. § 1.192

Sir:

This Appeal Brief is filed in triplicate pursuant to the *Notice of Appeal to the Board of Patent Appeals and Interferences* mailed October 20, 2003.

REAL PARTY IN INTEREST

The real party in interest is assignee Cree, Inc., Durham, North Carolina.

RELATED APPEALS

Appellant is aware of no appeals or interferences that would be affected by the present appeal.

STATUS OF CLAIMS

Claims 1-5, 7-24, 26-33, 35-36 and 46-48 remain pending. Each of these claims currently stands finally rejected. Appellants appeal the final rejection of Claims 1-5, 7-24, 26-33, 35-36 and 46-48. The attached Appendix A presents the claims at issue as finally rejected in the Official Action of July 8, 2003 and the Advisory Action of October 9, 2003.

STATE OF AMENDMENTS

The attached Appendix A presents the claims as they currently stand. Each of the claims remains as filed or as amended in the Amendment dated May 1, 2003. All amendments have been entered in the present case.

SUMMARY OF THE INVENTION

Pursuant to embodiments of the present invention, methods of growing silicon carbide crystals using electric arc sublimation techniques are provided. (Specification at 5, lines 1-2). Sublimation refers to a process whereby a material is converted from a solid state directly into a gaseous state and thereafter the resulting gas vapors are condensed back into a solid form. (See Webster's New World Dictionary at 1418, Appendix B). In embodiments of the present invention, at least one silicon carbide electrode may be placed in a sublimation system. (Specification at 5, lines 2-8). An electric arc may then be created between the silicon carbide electrode and a second electrode that raises the temperature of the silicon carbide electrode adjacent the arc to a temperature sufficient to sublime the silicon carbide. (Specification at 5, lines 2-8). In other embodiments of the invention, a localized heat source may be used instead of the electric arc to raise at least a portion of the silicon carbide source material to a temperature at which it sublimates. (Specification at 5, lines 8-10).

The power dissipation across the electric arc may be controlled to create a constant flow of vaporized Si, Si₂C and/or SiC₂ from the silicon carbide source independent of both the internal temperature and/or pressure of the sublimation system. (Specification at 5, lines 11-15). This may be accomplished, for example, by adjusting the position of one or both electrodes as the electric arc consumes the end of the electrodes so as to maintain a constant gap between the electrodes over time. (See, e.g., Specification at 10, lines 21-29). By using the power dissipation level across the electric arc as a means for maintaining a constant flux, the temperature and pressure of the sublimation system may be independently set to grow a desired polytype of silicon carbide. (Specification at 11, lines 19-29). This differs from conventional silicon carbide sublimation techniques where the pressure and/or temperature may be adjusted throughout the process in an effort to maintain a relatively constant source-to-seed flux of the vaporized silicon carbide. (Specification at 11, lines 19-29).

ISSUES

1. Are Claims 1-5, 7-12, 14-17, 20, 23-24, 26-33, 35-36 and 46-47 properly rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov (U.S. Patent No.

4,978,556) in view of Davis (U.S. Patent No. RE 34,861) and Kuehnle (U.S. Patent No. 5,879,518)?

2. Are Claims 1-5, 7-12, 14, 17-20, 23-24, 26-30, 32-33, 35-36 and 46-47 properly rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis and Smalley (U.S. Patent No. 5,227,038)?

3. Is Claim 13 properly rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Jaussaud (U.S. Patent No. 6,113,692), and Kuehnle or Smalley?

4. Are Claims 18-19 properly rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Kuehnle and Fey (U.S. Patent No. 4,582,004)?

5. Are Claims 21-22 properly rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Kuehnle or Smalley and Otsuki (U.S. Patent No. 6,090,733)?

6. Are Claims 22 and 48 properly rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Kuehnle or Smalley and Kijima (U.S. Patent No. 5,093,039)?

GROUPING OF CLAIMS

Claims 1-5, 7-24, 26-33, 35-36 and 46-48 stand rejected as obvious under 35 U.S.C. § 103. For the purposes of this appeal, Appellant submits that Claims 1-5, 7-10, 13, 16-24, 26-29, 31-32 and 46-48 may be considered as standing or falling together (Group I), Claims 33, 35 and 36 may be considered as standing or falling together (Group II), Claims 14 and 30 may be considered as standing or falling together (Group III), Claims 11 and 15 may be considered as standing or falling together (Group IV) and Claim 12 may also be considered as standing or falling independently of the remaining claims (Group V). Appellant submits that the above-listed groups of claims are separately patentable for the reasons discussed below.

ARGUMENT

I. INTRODUCTION

Each of the claims of the present application stands rejected as obvious under 35 U.S.C. § 103. A determination under Section 103 that an invention would have been obvious to someone of ordinary skill in the art is a conclusion of law based on fact. *Panduit Corp. v. Dennison Mfg. Co.* 810 F.2d 1593, 1 U.S.P.Q.2d 1593 (Fed. Cir. 1987), *cert. denied*, 107 S.Ct. 2187. After the involved facts are determined, the decision maker must then make the legal determination of whether the claimed invention as a whole would have been obvious to a person having ordinary skill in the art at the time the invention was unknown, and just before it was made. *Id.* at 1596. The United States Patent and Trademark Office has the initial burden under Section 103 to establish a *prima facie* case of obviousness. *In re Fine*, 837 F.2d 1071, 5 U.S.P.Q.2d 1596, 1598 (Fed. Cir. 1988).

To establish a *prima facie* case of obviousness, the prior art references cited in the rejection, when combined, must teach or suggest **all** the recitations of the claims, and there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to combine the reference teachings in the manner suggested. M.P.E.P. § 2143. The mere fact that references can be combined or modified does not render the resultant combination obvious unless the prior art also suggests the desirability of the combination. M.P.E.P. § 2143.01, citing *In re Mills*, 916 F.2d 680, 16 U.S.P.Q.2d 1430 (Fed. Cir. 1990). As emphasized by the Court of Appeals for the Federal Circuit, to support combining references, evidence of a suggestion, teaching, or motivation to combine must be clear and particular, and this requirement for clear and particular evidence is not met by broad and conclusory statements about the teachings of references. *In re Dembiczak*, 50 U.S.P.Q.2d 1614, 1617 (Fed. Cir. 1999). Thus, in support of a Section 103 rejection, particular evidence from the prior art must be provided showing why a skilled artisan, with no knowledge of the claimed invention, would have combined the cited references in the manner claimed in the rejection. *In re Kotzab*, 55 U.S.P.Q.2d 1313, 1317 (Fed. Cir. 2000).

Furthermore, as recently stated by the Federal Circuit with regard to the selection and combination of references:

This factual question of motivation is material to patentability, and could not be resolved on subjective belief and unknown authority. It is improper, in determining whether a person of ordinary skill would have been led to this combination of references, simply to "[use] that which the inventor taught against its teacher." *W.L. Gore v. Garlock, Inc.*, 721 F.2d 1540, 1553, 220 USPQ 303, 312-13 (Fed. Cir. 1983). Thus the Board must not only assure that the requisite findings are made, based on evidence of record, but must also explain the reasoning by which the findings are deemed to support the agency's conclusion....

In re Sang Su Lee, 277 F.3d 1338, 1343 (Fed. Cir. 2002).

Appellant respectfully submits that the pending claims are patentable over the cited references because the cited combination fails to disclose or suggest all of the recitations of the pending claims, and because the reasoning behind such combination has not been established. The patentability of the pending claims is discussed in detail hereinafter.

II. THE GROUP I-V CLAIMS ARE PATENTABLE OVER THE CITED ART

Claims 1-5, 7-12, 15-17, 20, 23-24, 26-29, 31-32 and 46-47 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis and Kuehnle. (Final Action at ¶ 2, pp. 2-5). Claims 1-5, 7-12, 17-20, 23-24, 26-29, 32, and 46-47 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis and Smalley. (Final Action at ¶ 3, pp. 5-7). Claim 13 stands rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Jaussaud, and Kuehnle or Smalley. (Final Action at ¶ 4, pp. 7-8). Claims 18-19 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Kuehnle and Fey. (Final Action at ¶ 5, pp. 8-9). Claims 21-22 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Kuehnle or Smalley and Otsuki. (Final Action at ¶ 6, pp. 9-10). Claims 22 and 48 stand rejected under 35 U.S.C. § 103(a) as being unpatentable over Pinkahsov in view of Davis, Kuehnle or Smalley and Kijima. (Final Action at ¶ 7, pp. 10-11). For the reasons discussed below, Appellant respectfully submits that all of these claims are patentable over the cited art.

A. The Rejections Should Be Reversed Because Pinkahsov Does Not Teach Growing Silicon Carbide Via Sublimation

The present application includes three independent claims, Claims 1, 24 and 33. For the Court's convenience, each of these claims are reproduced below:

1. A method of growing silicon carbide, the method comprising:
introducing a seed of silicon carbide into a sublimation system;
introducing a silicon carbide electrode into the sublimation system;
introducing a second electrode into the sublimation system adjacent the silicon carbide electrode, wherein the silicon carbide electrode and the second electrode are separated by a gap;
establishing an electric arc across the gap between the silicon carbide electrode and the second electrode to vaporize at least part of the silicon carbide electrode and cause at least some of the vaporized silicon carbide materials to form silicon carbide on the silicon carbide seed; and
controlling the power dissipated across the gap to control a flow of vaporized Si, Si₂C and SiC₂ from the silicon carbide electrode to the seed of silicon carbide.

24. A method of growing silicon carbide, the method comprising:
establishing an electrical arc between a pair of electrodes spaced apart by a gap in order to electrically arc a silicon carbide source to sublimate silicon and carbon containing material from the silicon carbide source and cause at least some of the silicon and carbon containing material to form silicon carbide on a silicon carbide seed; and
controlling the power dissipated across the gap to control the flow of vaporized Si, Si₂C and SiC₂ from the silicon carbide source to the silicon carbide seed.

33. A method of growing silicon carbide, the method comprising:
using resistive or inductive heating to heat a furnace to a temperature below the temperature at which silicon carbide sublimates;
using an electric arc to create a local high temperature zone within a the furnace that is above the temperature at which silicon carbide sublimates while maintaining the inner walls of the furnace at a temperature below the temperature at which silicon carbide sublimates;
introducing a silicon carbide source material into the high temperature zone to sublimate silicon and carbon containing material from the silicon carbide source and cause at least some of the silicon and carbon containing material to form silicon carbide on a silicon carbide seed.

As is clear from the above, all of the pending claims are directed to methods of growing silicon carbide via electric arc sublimation. (See, e.g., Claim 1, stating that a seed of silicon carbide is introduced "into a sublimation system"; see also Claims 24 and 33, stating that an electrical arc is used "to sublimate silicon and carbon." The Advisory Action states that the Pinkahsov reference is relied upon as teaching the electric arc sublimation of silicon carbide recitations of the pending claims. (See Advisory Action at 2).

Appellant respectfully submits that the rejections of all of the pending claims should be withdrawn because Pinkahsov does not teach sublimating silicon carbide using electric arc vapor deposition techniques. Sublimation refers to a process where a solid is heated directly to a gaseous state and the resulting vapors are condensed back into solid form. (See Webster's Dictionary at 1418, defining sublimate and sublime, attached as Appendix B hereto). Pinkahsov, however, is directed to an arc vapor deposition technique in which molten (i.e., material that has been "melted or liquefied by heat") material is vaporized for purposes of growing a material on a substrate. (See Pinkahsov at Col. 2, lines 34-36; Col. 3, lines 49-54; see also Webster's Dictionary at 916, Appendix B, defining molten). Evaporating a molten liquid, by definition, is not sublimation. Thus, as none of the cited references teach or suggest growing silicon carbide via electric arc sublimation the rejections should be reversed.

In the Advisory Action, the Examiner argues that the Pinkahsov reference in fact teaches sublimation and is not limited to embodiments that "require a molten pool." (See Advisory Action at 2, citing to Pinkahsov at Col. 1, lines 34-43 and Claim 5). However, the portion of Pinkahsov cited by the Examiner is a summary of arc-vapor deposition techniques that are disclosed in a series of prior patents to Pinkahsov. (See Pinkahsov at Col. 1, lines 29-43). Pinkahsov expressly states that it is these prior patents that describe the details regarding "[t]he formation of vapor and its deposition upon the substrate." (Pinkahsov at Col. 3, lines 54-57). These prior patents to Pinkahsov make clear that the Pinkahsov process involves two phase transformations – solid to liquid and liquid to gas.

For instance, one of the prior patents that is summarized in Pinkahsov describes the arc vapor deposition technique as follows:

[A] body of the material to be transferred to the substrate is heated in the region of this substrate and transformed first into a molten state and then into a vapor state. The material thus undergoes two phase transformations, namely, the transformation from the solid phase to the liquid phase and then from the liquid phase to the vapor phase.

(U.S. Patent No. 4,505,948 to Pinkahsov at Col. 1, lines 42-48). The prior patents go on to explain that the vapor deposition proceeds by striking an electric arc between the pool of molten material and an electrode:

This application discloses a method of vapor-depositing material upon a substrate which, as indicated, utilizes an electrical arc struck between a pool of molten

material and a counterelectrode, thereby vaporizing the material on the surface of the pool and permitting transfer of the vaporized material in the vapor state to the substrate.

(*Id.* at Col. 2, lines 13-17). The Pinkahsov patent cited by the Examiner likewise confirms that it relates to a process in which molten materials are vaporized by repeatedly stating that the arc is applied to a **molten electrode material**. (Pinkahsov at Col. 2, lines 31-36 and Col. 3, lines 49-54).

In summary, the portions of Pinkahsov relied upon by the Examiner to support the pending rejections do not teach growing silicon carbide via electric arc **sublimation** as recited in all of the pending claims, but instead disclose a two-step vaporization process that does not involve sublimation. The Examiner cannot change this fact by citing to sentences in Pinkahsov that describe the vaporization process in summary fashion as a teaching that sublimation could be used instead of a two-step process. As such, the rejections of all the pending claims should be reversed.¹

B. Pinkahsov Does Not Enable Arc-Vapor Deposition Using a Silicon Carbide Electrode

The rejections of the pending claims should likewise be reversed because Pinkahsov does not **enable** a person of skill in the art to perform electric arc vapor deposition using a silicon carbide electrode. As noted above, Pinkahsov discloses an arc vapor-deposition method in which an arc is struck between a molten pool of electrode material and a second electrode. Pinkahsov states that "the electrode material is crystalline silicon or silicon carbide or silicon nitride." (Pinkahsov at Col. 2, lines 47-48). However, the example in Pinkahsov only describes use of two **silicon** electrodes. Pinkahsov does further state that the manner in which the vapor is formed is fully described in a series of

¹ The Advisory Action also states that "Smalley et al teaches a method of electric arc processing, where electrode material is vaporized without forming a molten pool." (Advisory Action at 2). Smalley, however, relates to an electric arc process that uses a different material system for a completely different purpose, namely generating fullerenes by vaporizing carbon. (*See* Smalley at Abstract). The Examiner has not even attempted to explain why a skilled artisan would have been motivated to modify the process of Pinkahsov to operate via sublimation based on Smalley, and Appellant respectfully submits that the only such motivation comes from hindsight and using the present invention as a roadmap to piece together various teachings from a wide variety of prior art references.

prior, related patents. (Pinkahsov at Col. 1, lines 29-34 and Col. 3, lines 54-57). These prior patents, however, explain that the silicon carbide is deposited on the substrate by using a silicon electrode and a carbon block electrode which contains "a molten pool of silicon and solubilized carbon." (See U.S. Patent No. 4,505,948 to Pinkahsov at Col. 6, lines 60-63 and Col. 7, lines 36-40). Thus, Pinkahsov does not explain how the process could be carried out using a silicon carbide electrode.

This failure of Pinkahsov is significant because it is clear that the process of Pinkahsov **would not work** with a silicon carbide electrode. Pinkahsov teaches that the process is carried out under vacuum conditions (*i.e.*, at pressures of 10^{-3} torr or better). (Pinkahsov at Col. 3, lines 66-68). As shown in the references attached at Appendix C hereto, silicon carbide **does not form a liquid state at those pressures** (See, *e.g.*, Seace and Slack, Solubility of Carbon in Silicon and Germanium at 1554). Accordingly, the method of Pinkahsov where the arc vapor deposition is carried out between an electrode and a pool of molten material in a second electrode will not work with silicon carbide electrodes. Thus, because Pinkahsov does not enable an arc vapor-deposition process that uses a silicon carbide electrode, the rejections of all the pending claims should be withdrawn.

C. The Rejections Should Be Reversed Because There is No Motivation To Combine Kuehnle or Smalley with Pinkahsov and Davis

As noted above, to establish a *prima facie* case of obviousness, the combination of prior art references must teach or suggest all the recitations of the claims **and** there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to combine the reference teachings to arrive at the claimed invention. M.P.E.P. § 2143. The mere fact that references can be combined or modified does not render the resultant combination obvious unless the prior art also suggests the desirability of the combination.

All of the pending claims stand rejected under 35 U.S.C. § 103 based, at least in part, on (1) the combination of Pinkahsov, Davis and Kuehnle and (2) the combination of Pinkahsov, Davis and Smalley. In these rejections, the Examiner relies on Kuehnle or Smalley for teaching the step of "controlling the power dissipated across the gap to control

a flow of vaporized Si, Si₂C and SiC₂ from the silicon carbide electrode to the seed of silicon carbide." (See, e.g., Final Office Action at ¶ 2, p. 4). The Final Office Action, however, fails to provide the necessary showing that a skilled artisan would have been motivated to combine this alleged teaching of either Kuehnle or Smalley with Pinkahsov and Davis. The lack of any such evidence of motivation to combine the references in the manner of the rejections provides an independent basis for withdrawal of each of the pending rejections.

The portion of the Final Office Action detailing the rationale for the pending rejections conclusively states that "[i]t would have been obvious to a person of ordinary skill in the art at the time of the invention to modify the combination of Pinkahsov and Davis et al with Kuehnle to vaporize the electrode material at a steady level (col. 4, ln 15-25), thereby avoiding undesired changes in flux resulting in uniform polytype." (Final Office Action at 4). With respect to the rejections that rely on Smalley, the Final Office Action similarly states in a conclusory fashion that "[i]t would have been obvious to a person of ordinary skill in the art at the time of the invention to modify the combination of Pinkahsov and Davis et al with Smalley's means for maintaining a desired gap for the electrical arc to maintain an optimum length of the arc gap during the entire process." (Final Office Action at 6). In the Advisory Action, the Examiner argues that the teaching in Davis that the silicon carbide should be vaporized at a constant flux provides the motivation for combining Kuehnle and/or Smalley with Pinkahsov and Davis. (Advisory Action at 3). However, Appellant respectfully submits that a skilled artisan would not combine the references as suggested in the pending rejections for at least two independent reasons.

First, neither Kuehnle nor Smalley relate to depositing silicon carbide on a substrate via electric arc sublimation. Kuehnle is directed to evaporating particles at a steady state so that the particles will cluster together at a consistent rate. (See Kuehnle at Col. 4, lines 13-23). Smalley is directed to a method of forming fullerenes, a molecular form of carbon. (See Smalley at Abstract). Thus, the suggestions in both Kuehnle and Smalley regarding the gap to maintain between the electrodes is made in the context of a different art and directed to solving a different problem. **The Davis reference, however, contains explicit teachings regarding controlling the flux between the silicon carbide**

source material and the silicon carbide seed. In particular, Davis teaches that the "if the thermal gradient is continually increased as the source powder is depleted and as the seed crystal grows, an absolute temperature differential between the source and the seed can be maintained at an amount which continues to be most favorable for growth." (Davis at Col. 9, lines 29-35). Davis further teaches that this can be accomplished by modifying the temperature of the source powder and/or the seed during growth and/or by changing the pressure during growth or by a combination of these methods. Appellant respectfully submits that the skilled artisan would look to the teachings in the Davis references regarding controlling the temperature and the pressure in the reactor to maintain a steady flux or the teachings of some other reference that actually relates to the field of the present invention. What the skilled artisan would not do is look to a completely different field to directly modify the teachings of a reference that the Final Office Action contends teaches the method of solving the problem addressed by the present invention. Thus, the pending rejections should be withdrawn because the Examiner has not and cannot show that a skilled artisan would have combined Pinkashov and Davis with either Kuehnle or Smalley in the manner suggested.

The combinations of references relied upon in the rejections is also unsupportable because **Pinkahsov teaches directly away from using the electrode apparatus disclosed in Kuehnle or Smalley.** In particular, Pinkahsov teaches that "arc vapor deposition" of silicon or silicon carbide onto a substrate is accomplished by repeatedly striking two electrodes together and then separating the electrodes to form an arc between them. (Pinkahsov at Col. 1, lines 34-45 and Co. 3, lines 49-60). In the example set forth in Pinkahsov, an arc is formed approximately one time per second through this intermittent movement of one or both of the electrodes. (Pinkahsov at Col. 4, lines 27-28). As explained in one of the earlier patents that Pinkahsov references as describing aspects of his invention:

I may move the counterelectrode into and out of contact with the pool to thereby deposit some of the melt upon the counterelectrode and permitting the heat generated at the electrode tip to vaporize at least in part the material transferred to it and thus in part generate the vapors which are to be transferred to the substrate.

(U.S. Patent No. 5,505,948 at Col. 2, lines 30-36). This prior Pinkahsov patent goes on to state:

Surprisingly, once the arc is struck as the two electrodes are separated, the arc, a portion of the arc or a heating effect generated by the arc appears to spiral around the long electrode and cause vaporization of the material of the electrode in a generally helical or spiral pattern progressively moving away from the counterelectrode.

It is indeed a remarkable surprise that the arc is not confined to the space between the two electrodes but rather has a component or an effect which spirals away from the counterelectrode toward a region of the length of the long electrode which is further removed from the counterelectrode in spite of the fact that the greatest conductivity would appear to lie in a line directly between the two electrodes where the major portion of the arc appears to be confined. This effect is manifest in the fact the long electrode, i.e. the deposition electrode, while originally of uniform cross section, develops a taper toward the counterelectrode and coating from the blank of the deposition electrode onto the substrate can be observed at considerable distance from the arc's striking face of the deposition electrode.

In fact, the effect appears to survive for a brief period following extinction of the original arc and hence I prefer to periodically contact and separate the electrodes to generate the arc and then allow extinction thereof.

(U.S. Patent No. 5,505,948 at Col. 4, lines 16-42). Thus, it is clear that a critical part of the invention of Pinkahsov involves moving the electrodes into and out of contact with each other – in other words, **not** maintaining a constant gap – as this is required both to (1) facilitate vaporization of the electrode material in the molten pool by depositing that material onto the tip of the counterelectrode and (2) to induce an effect whereby the arc is formed not only between the two electrodes, but also along the length of the deposition electrode.

In light of these teachings in Pinkahsov, Appellant respectfully submits that a skilled artisan would not have been motivated to combine either Kuehnle or Smalley with Pinkahsov as doing so would be **directly contrary** to the teachings of Pinkahsov.

Appellant notes that in the Advisory Action the Examiner argues that Pinkahsov is not limited to embodiments in which the electrodes are struck together to form the arc.

However, what the Examiner has not and cannot do is point to any other arc vaporization process in Pinkahsov, because the only process disclosed is one in which the electrodes are struck together. In fact, the very portion of Pinkahsov that the Examiner cites to as teaching that Pinkahsov is not limited to embodiments in which the electrodes are struck together expressly states that the electrodes are brought into contact and separated to form

the arc. (*See* Advisory Action at 3-4, citing to Pinkahsov at Col. 1, lines 29-45). As such, Appellant respectfully submits that this teaching away from the combination of references cited in the pending rejections compels reversal of those rejections.

D. The Rejections Should Be Withdrawn Because Kuehnle and Smalley are Not Analogous Art

Finally, the pending rejections of the Group I-V claims should also be withdrawn because neither Kuehnle nor Smalley can properly be relied upon as part of a rejection under 35 U.S.C. § 103. In particular, only references that come from an "analogous art" may be relied upon to support a rejection under Section 103, where a reference is considered to be from an analogous art if it either (1) is from the same field of endeavor as the invention at issue, regardless of the problem addressed or (2) is reasonably pertinent to the particular problems with which the inventor is involved. *See, e.g., In re Paulsen*, 31 U.S.P.Q.2d 1671, 1675-76 (Fed. Cir. 1994). Neither Kuehnle nor Smalley satisfy this test.

As to the first prong of the analogous art test, the present invention is directed to the growth of semiconductor materials on a seed crystal or substrate. In contrast, Kuehnle is directed to "a method for producing small particles, e.g., nanoparticles, which have consistent size, shape, structure and functionality." (Kuehnle at Col. 1, lines 7-10). Smalley likewise has nothing to do with growing semiconductor materials on a substrate, but instead is directed to a method for making fullerenes (a particular form of carbon). (*See* Smalley at Abstract). Thus, both Kuehnle and Smalley are from different fields of endeavor, and hence neither reference qualifies as analogous art under the first prong of the test.

The second prong of the analogous art test examines whether or not a reference is reasonably pertinent to the particular problems with which the invention is involved. Here, that problem is growing monocrystalline and/or polycrystalline silicon carbide crystals on a substrate for use in semiconductor applications. Kuehnle has nothing to do with the problems which the present invention addresses, as Kuehnle provides insights regarding how to collect a powder of a source substance where the particles of the powder are relatively uniform in size. Smalley is directed to a method of growing fullerenes – a form of carbon – that has no semiconductor applications. Thus, neither Kuehnle nor Smalley qualify as analogous art under the second prong of the test and, as such, the

claims of the present application cannot properly be rejected based on the combination of the Pinkashov and Davis and either Kuehnle or Smalley.

III. THE GROUP II CLAIMS ARE PATENTABLE OVER THE CITED ART

The Group II claims are Claims 33, 35 and 36. The Group II claims stand rejected under 35 U.S.C. § 103 based on the combination of (1) Pinkashov, Davis and Kuehnle and (2) Pinkashov, Davis and Smalley. (*See* Final Office Action at ¶¶ 2, 3, pp. 2-7). Appellant respectfully submits that each of the Group II claims are patentable over the cited references for each of the reasons set forth in Section II above. In addition, Appellant submits that the Group II claims are also patentable over the cited art for at least one additional reason.

Each of the Group II claims recite that the electric arc is used to "create a local high temperature zone within the furnace . . . while maintaining the inner walls of the furnace at a temperature below the temperature at which silicon carbide sublimates." Appellant respectfully submits that none of the cited references teach or suggest "using resistive or inductive heating to heat a furnace" while at the same time "maintaining the inner walls of the furnace at a temperature below the temperature at which silicon carbide sublimates." (*See* Claim 33). Appellant raised the above argument in Appellant's Response to Final Office Action. In response, the Examiner argued in the Advisory Action that the Group II claims do "not require heating the inner walls of the furnace." (Advisory Action at 4). The Examiner, however, **apparently overlooked** the recitation of Claim 33 that recites "using resistive or inductive heating to heat a furnace to a temperature below the temperature at which silicon carbide sublimates." Heating the furnace includes heating the inner walls of the furnace. Thus, as it is clear that the rejections of the Group II claims are based on a mistake by the Examiner regarding the actual elements included within the Group II claims, Appellant respectfully submits that the rejections of the Group II claims should be reversed.

In any event, Appellant submits that:

- Davis teaches heating the furnace walls to a temperature **above** the temperature at which silicon carbide sublimates. (*See, e.g.,* Davis at Col. 11, lines 20-23).

- Pinkahsov does not teach or suggest that a furnace is even provided, as the electric arc is used solely to perform the vaporization. (See Pinkahsov at Col. 4, lines 5-41).
- Smalley does not teach or suggest that a furnace is even provided, as the electric arc is used solely to perform the vaporization. (See Smalley at Col. 2, lines 46-48).
- Kuehnle teaches that a "cold" inert gas is flowed just inside the housing sidewall, indicating that in Kuehnle the housing is actually cooled as opposed to heated. (See Kuehnle at Col. 4, lines 3-12).

This showing that the cited art does not disclose each of the recitations of the Group II claims is un rebutted. Accordingly, Appellant respectfully requests that the rejections of the Group II claims be reversed for this additional reason.

IV. THE GROUP III CLAIMS ARE PATENTABLE OVER THE CITED ART

The Group III claims are Claims 14 and 30. Appellant respectfully submits that each of the Group III claims are patentable over the cited references for the reasons set forth in Section II above. In addition, Appellant submits that the Group III claims are also patentable over the cited art for at least one additional reason.

Claim 14 recites that "the internal temperature of the sublimation system, the position of the silicon carbide electrode and the second electrode, a voltage drop across the gap and a current conducted across the gap are configured so as to maintain the end of the silicon carbide electrode adjacent the gap at a substantially constant temperature during the sublimation process." Claim 30 contains a similar recitation. While the Final Office Action states that the combination of Pinkahsov and Davis in view of either Kuehnle or Smalley teaches these recitations, Appellant respectfully disagrees. Pinkahsov discloses a process where the temperature at the end of the electrode will vary (*i.e.*, not remain constant), and Davis teaches that the temperature of the silicon carbide source material is varied throughout the sublimation process. Kuehnle and Smalley are silent regarding the temperature at the end of the electrode, and hence do not provide the necessary teaching. In any event, even if Kuehnle or Smalley taught the recitations of the Group III claims, no motivation has been identified for modifying the primary references – each of which teach away from maintaining the ends of the electrodes at a constant temperature – in this

manner. Accordingly, the Group III claims likewise are independently patentable over the cited art for at least these additional reasons.

In the Advisory Action, the Examiner takes the position that the recitations of Claims 14 and 30 are "recitation[s] of the intended use of the claimed invention," and hence cannot patentably distinguish the invention from the prior art. (Advisory Action at 5). However, the recitations at issue simply are not recitations of the intended use of the invention, but instead comprise one of the conditions that must be met to practice the claimed methods. Thus, as each of the recitations of the Group III claims are not taught or suggested by the cited references, the rejections of the Group III claims should be reversed.

V. THE GROUP IV CLAIMS ARE PATENTABLE OVER THE CITED ART

The Group IV claims are Claims 11 and 15. Appellant respectfully submits that each of the Group IV claims are patentable over the cited references for the reasons set forth in Section II above. In addition, Appellant submits that the Group III claims are also patentable over the cited art for at least one additional reason.

Each of the Group IV claims recite that a "substantially constant pressure level is pre-selected for growth of a pre-selected polytype of silicon carbide." The Advisory Action takes the position that Davis teaches this recitation at Column 13, lines 1-25. Appellant respectfully submits that this is not the case. Specifically, the cited portion of Davis states that the furnace was initially maintained at a pressure of 400 Torr during the growth process. Then, over a period of 85 minutes, the system was slowly evacuated to reduce the pressure from 400 Torr to 10 Torr. The pressure remained at 10 Torr for 6 hours, and then the pressure was increased to 760 Torr. Thus, the cited portion of Davis teaches that the silicon carbide was grown under a variety of different pressure conditions, and clearly does not teach or suggest pre-selecting a pressure level for growth of a pre-selected polytype of silicon carbide. Accordingly, the rejections of the Group IV claims should also be reversed.

VI. THE GROUP V CLAIM IS PATENTABLE OVER THE CITED ART

The Group V claim is Claim 12. Appellant respectfully submits that Claim 12 is patentable over the cited references for the reasons set forth in Section II above. In addition,

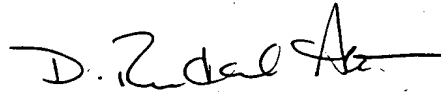
Appellant submits that Claim 12 is also patentable over the cited art for at least one additional reason.

Claim 12 recites "raising the temperature of the silicon carbide electrode to a temperature lower than the temperature at which silicon carbide sublimes." Neither the Final Office Action nor the Advisory Action even attempt to explain where this recitation may be found in the cited references. Davis clearly does not teach it, as Davis does not disclose an electrode and, in any event, teaches heating the silicon carbide source material to a temperature above the temperature at which silicon carbide sublimes. Pinkashov, Kuehnle and Smalley do not teach raising the temperature of the silicon carbide electrode at all, aside from the temperature increase that is caused by the electric arc vaporization, and that temperature increase is, again, to a temperature above the temperature at which silicon carbide sublimes. Accordingly, Claim 12 is also independently patentable over the cited art for at least these additional reasons.

V. CONCLUSION

In light of the above discussion, Appellant submits that each of the pending claims is patentable over the cited references and, therefore, request reversal of the rejections of Claims 1-5, 7-24, 26-33, 35-36 and 46-48.

Respectfully submitted,

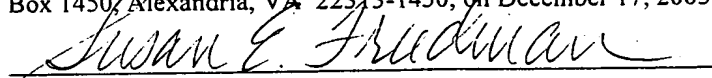


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I hereby certify that this correspondence is being deposited with the United States Postal Service as first class mail in an envelope addressed to: Mail Stop Appeal-Brief Patents, Commissioner for Patents, P.O. Box 1450, Alexandria, VA 22313-1450, on December 17, 2003.



Susan E. Freedman

Date of Signature: December 17, 2003

APPENDIX A
Pending Claims USSN 09/931,537
Filed August 16, 2001

1. (Previously Amended): A method of growing silicon carbide, the method comprising:

introducing a seed of silicon carbide into a sublimation system;

introducing a silicon carbide electrode into the sublimation system;

introducing a second electrode into the sublimation system adjacent the silicon carbide electrode, wherein the silicon carbide electrode and the second electrode are separated by a gap;

establishing an electric arc across the gap between the silicon carbide electrode and the second electrode to vaporize at least part of the silicon carbide electrode and cause at least some of the vaporized silicon carbide materials to form silicon carbide on the silicon carbide seed; and

controlling the power dissipated across the gap to control a flow of vaporized Si, Si₂C and SiC₂ from the silicon carbide electrode to the seed of silicon carbide.

2. (Original): The method of Claim 1, wherein the second electrode is a silicon carbide electrode.

3. (Previously Amended): The method of Claim 2, wherein controlling the power dissipated across the gap further controls a flow of vaporized Si, Si₂C and SiC₂ from the second electrode to the seed of silicon carbide.

4. (Previously Amended): The method of Claim 3, wherein controlling the power dissipated across the gap comprises moving at least one of the silicon carbide electrode and the second electrode as they vaporize during the sublimation process to maintain a constant gap between the silicon carbide electrode and the second electrode.

5. (Original): The method of Claim 2, further comprising maintaining the pressure within the sublimation system at a substantially constant level during the sublimation process.

6. (Cancelled)

7. (Previously Amended): The method of Claim 1, wherein the power dissipated across the gap is controlled to maintain a substantially constant flow of vaporized Si, Si₂C and SiC₂ per unit area per unit time from the silicon carbide electrode to the seed crystal.

8. (Previously Amended): The method of Claim 1, wherein controlling the power dissipated across the gap comprises moving at least one of the silicon carbide electrode and the second electrode during the sublimation process to maintain a constant gap between the silicon carbide electrode and the second electrode.

9. (Original): The method of Claim 1, further comprising moving at least one of the silicon carbide electrode and the second electrode to maintain a substantially constant separation between the silicon carbide electrode and the second electrode.

10. (Original): The method of Claim 1, further comprising maintaining the pressure within the sublimation system at a substantially constant level during the sublimation process.

11. (Previously Amended): The method of Claim 10, where the substantially constant pressure level is set to ensure that a specific polytype of silicon carbide is grown.

12. (Original): The method of Claim 10, further comprising:
raising the temperature of the seed to a temperature lower than the temperature at which silicon carbide sublimates; and
raising the temperature of the silicon carbide electrode to a temperature lower than the temperature at which silicon carbide sublimates.

13. (Previously Amended): The method of Claim 12, wherein the sublimation system includes a furnace, and wherein the method further comprises raising the temperature of the inner walls of the furnace to a temperature higher than the temperature of the seed.

14. (Previously Amended): The method of Claim 1, wherein the internal temperature of the sublimation system, the position of the silicon carbide electrode and the second electrode, a voltage drop across the gap and a current conducted across the gap are configured

so as to maintain the end of the silicon carbide electrode adjacent the gap at a substantially constant temperature during the sublimation process.

15. (Previously Amended): The method of Claim 14, where the substantially constant temperature is set to ensure that a specific polytype of silicon carbide is grown.

16. (Previously Amended): The method of Claim 1, wherein controlling the power dissipated across the gap to control a flow of vaporized Si, Si₂C and SiC₂ from the silicon carbide electrode to the seed crystal comprises:

sensing a voltage drop across the gap; and

adjusting the relative location of the silicon carbide electrode and the second electrode so as to maintain the voltage drop at a constant level.

17. (Original): The method of Claim 16, further comprising rotating the seed during at least part of the sublimation process.

18. (Previously Amended): The method of Claim 1, wherein establishing an electric arc between the silicon carbide electrode and the second electrode comprises activating an alternating current power supply that is electrically connected to one of the silicon carbide electrode and the second electrode.

19. (Original): The method of Claim 18, wherein the frequency at which the alternating current power supply is operated is selected to maintain substantially the same rate of vaporization of the silicon carbide electrode.

20. (Original): The method of Claim 2, wherein the silicon carbide electrode is formed by sintering silicon carbide powder.

21. (Original): The method of Claim 20, wherein the silicon carbide electrode is formed from an n-type carrier rich silicon carbide source powder.

22. (Original): The method of Claim 20, wherein the silicon carbide electrode is formed from a p-type carrier rich silicon carbide source powder.

23. (Previously Amended): The method of Claim 2, wherein the internal temperature of the sublimation system, the pressure within the sublimation system and the voltage and current associated with the electric arc are maintained so as to heat a constant volume of the silicon carbide electrode above the temperature where sublimation occurs during a crystal growth phase of the sublimation process.

24. (Previously Amended): A method of growing silicon carbide, the method comprising:

establishing an electrical arc between a pair of electrodes spaced apart by a gap in order to electrically arc a silicon carbide source to sublime silicon and carbon containing material from the silicon carbide source and cause at least some of the silicon and carbon containing material to form silicon carbide on a silicon carbide seed; and

controlling the power dissipated across the gap to control the flow of vaporized Si, Si₂C and SiC₂ from the silicon carbide source to the silicon carbide seed.

25. (Cancelled)

26. (Previously Amended): The method of Claim 24, wherein the electrodes are silicon carbide electrodes that serve as the silicon carbide source.

27. (Original): The method of Claim 26, wherein the power dissipated across the gap is controlled to maintain a substantially constant flow of vaporized Si, Si₂C and SiC₂ per unit area per unit time from the pair of silicon carbide electrodes to the silicon carbide seed.

28. (Original): The method of Claim 26, wherein controlling the power dissipated across the gap comprises moving at least one of the pair of silicon carbide electrodes as they vaporize during the sublimation process to maintain a constant gap between the pair of silicon carbide electrodes.

29. (Original): The method of Claim 24, further comprising maintaining the pressure within the sublimation system at a substantially constant level during the sublimation process.

30. (Previously Amended): The method of Claim 24, wherein the sublimation process occurs within a heated furnace, and wherein internal temperature of the furnace, the position of the pair of silicon carbide electrodes, the voltage drop across the spacing between the pair of electrodes and the arc current are configured so as to maintain the ends of the pair of electrodes adjacent the arc at a substantially constant temperature during the sublimation process.

31. (Original): The method of Claim 26, wherein controlling the power dissipated across the gap to control the flow of vaporized Si, Si₂C and SiC₂ from the pair of silicon carbide electrodes to the silicon carbide seed comprises:

sensing a voltage drop across the gap; and

adjusting the relative location of the silicon carbide electrodes so as to maintain the voltage drop at a constant level.

32. (Previously Amended): The method of Claim 24, wherein the sublimation process occurs within a heated furnace, and wherein an internal temperature of the furnace, a pressure within the furnace and the voltage and current associated with the arc are maintained so as heat a constant volume of the silicon carbide source above the temperature where sublimation occurs during a the crystal growth phase of the sublimation process.

33. (Previously Amended): A method of growing silicon carbide, the method comprising:

using resistive or inductive heating to heat a furnace to a temperature below the temperature at which silicon carbide sublimates;

using an electric arc to create a local high temperature zone within a the furnace that is above the temperature at which silicon carbide sublimates while maintaining the inner walls of the furnace at a temperature below the temperature at which silicon carbide sublimates;

introducing a silicon carbide source material into the high temperature zone to sublimate silicon and carbon containing material from the silicon carbide source and cause at

least some of the silicon and carbon containing material to form silicon carbide on a silicon carbide seed.

34. (Cancelled)

35. (Original): The method of Claim 33, wherein the silicon carbide source material is introduced into the high temperature zone by moving the silicon carbide source material.

36. (Previously Amended): The method of Claim 33, wherein the silicon carbide source material is introduced into the high temperature zone by moving a heating source used to create the local high temperature zone.

37. (Cancelled)

38. (Cancelled)

39. (Cancelled)

40. (Cancelled)

41. (Cancelled)

42. (Cancelled)

43. (Cancelled)

44. (Cancelled)

45. (Cancelled)

46. (Original): The method of Claim 1, wherein the silicon carbide seed is a monocrystalline seed of silicon carbide, and wherein the silicon carbide formed on the monocrystalline silicon carbide seed is monocrystalline silicon carbide.

47. (Original): The method of Claim 24, wherein the silicon carbide formed on the silicon carbide seed is monocrystalline silicon carbide.

48. (Original): The method of Claim 2, wherein the silicon carbide electrode is formed from silicon carbide powder grown by a chemical vapor deposition technique.

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< *L. molestare*: < *molestus*, troublesome < *moles*, a burden: see *MOL*. 1. to annoy, interfere with, or meddle with so as to trouble or harm, or with intent to trouble or harm 2. to make improper advances to, esp. of a sexual nature — *moles-ta-tion* (mō'les tā'shən, māl'ās-) *n.* — *mo-les-ter* *v.*

Mol-li-ère (mōl'yēr'; mō'lē'ēr'; Fr. mōl'yēr') (pseud. of Jean Baptiste Poquelin) 1622–73; Fr. dramatist

Mol-line (mōl'lēn') [*< Sp. molino*; *mull*] city in NW Ill., on the Mississippi; pop. 46,000

mo-line (mōl'in', mō'līn') *adj.* [*< Anglo-Fr. *moliné < OFr. molin*, a mill < VL. **molinum*, for LL. *molina*, *mull*; from its resemblance to the iron support for the upper millstone] designating a cross with each arm forked and curved back at the end

Moll (māl) a feminine name: see *MARY* — *n.* [usually *m-*] [Slang] 1. a gangster's mistress 2. a prostitute

mol-lāh (māl'ās) *n.* same as *MULLAH*

mol-les-cent (mōl'les'nt) *adj.* [L. *mollescens*, *pp.* of *mollescere*, to soften < *molle*, to be soft < *mollis*, soft; see *MOLLIFY*] softening or tending to soften — *mol-les-cence* *n.*

mol-lif-ty (māl'fī'tī) *v.* -fied, -fy'ing [ME. *molifen*; MFr. *mollifier* < LL. *mollificare*, to soften < L. *mollis*, soft (< IE. **mel-*, soft < base **mel-*, to crush, whence *MULL*) + *facere*, to make, do] 1. to soothe the temper of; pacify; appease 2. to make less intense, severe, or violent — *SYN.* see *PACIFY* — *mol-lif-ty-ca-tion* *n.* — *mol-lif-ty'er* *n.*

mol-lusc (māl'ask) *n.* same as *MOLLUSK* — *mol-lus-can* (mōl'us'kən) *adj.* *n.*

mol-lus-coid (mōl'us'koid) *adj.* 1. of or like a mollusk or mollusks 2. in some classifications, designating or of a group (Molluscoidea) of phyla comprising the brachiopods, phoronids, and bryozoans; all of which have a lophophore — *n.* a molluscoid animal

mol-lusk (māl'ask) *n.* [Fr. *mollusque* < ModE. *Mollusca*, coined by Cuvier < L. *mollusca*, a soft-shelled nut < *molluscus*, soft < *mollis*; see *MOLLIFY*] any of a large phylum (Mollusca) of invertebrate animals including the clitons, oysters, clams, mussels, snails, whelks, slugs, squids, octopuses, etc., characterized by a soft, usually unsegmented body, often enclosed wholly or in part in a mantle and a calcareous shell, and usually having gills and a foot — *mol-lus-kan* (mōl'us'kən) *adj.* *n.*

Moll-weide projection (mōl'vi'də) [after Karl B. Mollweide (1774–1825), G. mathematician] an equal-area map projection with the whole earth on one map, showing the prime meridian and all parallels of latitude as straight lines and all other meridians as increasing in curvature toward the margins

Mol-ly (māl'ē) a feminine name: see *MARY*

mo-l'y (māl'ē) *n.* *pl.* -lies [short for ModE. *Mollenisia*, genus name < F. N. Mollén (1758–1850), Fr. statesman] any of a genus (*Mollenisia*) of various brightly colored tropical and subtropical American fishes often kept in aquariums; also *mol'lie*

mol-ly-cod-dle (māl'ē kōd'ī) *n.* [*MOLLY* + *CODDLE*] a man or boy used to being coddled, or protected, pampered, etc.; milksop — *vt.* -died, -ding to pamper; coddle — *mol-ly-cod-dler* *n.*

Molly Ma-guires (mōl'gwi'z) [its members were sometimes disguised as women] 1. a secret society organized in Ireland in 1843 to prevent evictions by terrorizing agents of landlords 2. a secret society of Irish-American miners in E Pennsylvania (c. 1865–1875), which opposed oppressive industrial and social conditions, sometimes with physical force

Mol-nár (mōl'nār); Fe-renc (fe'rents) 1878–1932; Hung. playwright & novelist, later in the U.S.

Mo-loch (mōl'āk, māl'āk) same as *MOLECH* — *n.* (m-) a spiny-headed Australian lizard (*Moloch horridus*)

Mo-lo-kai (mō'lō'kī) [Haw. < ?] island of Hawaii, southeast of Oahu; site of a leper colony; 259 sq. mi.; pop. 6,000

Mo-lo-tov (mō'lō'tōf, V(yachestav) M(ikhalovich) (born Vyacheslav Mikhailovich Shriabin) 1890– : Russ. statesman; foreign minister of the U.S.S.R. (1939–49; 1953–56)

Molotov cocktail [after prec.] [Slang] a bottle filled with gasoline, etc. and wrapped in a saturated rag or plugged with a wick, ignited, and hurled as an antitank grenade

molt (mōlt) *vi.* [ME. *mouten* (with unhistoric *-t* after *FAULT*, in which the letter was orig. silent) < OE. (*be*) *mutian*, to exchange < L. *mutare*, to change; see *MUTATE*] to cast off or shed the hair, outer skin, horns, or feathers at certain intervals, prior to replacement of the castoff parts by a new growth; said of certain animals, as reptiles, birds, etc. — *vt.* to replace by molting — *n.* 1. the act or process of molting 2. the parts so shed — *molt'er* *n.*

mol-ten (mōl'tēn) [ME.] archaic *pp.* of *MELT* — *adj.* 1. melted or liquefied by heat 2. made by being melted and cast in a mold

Molt-ke (mōlt'kē) 1. Count Hel-muth (Johannes Ludwig) von (hel'mōt'fōn), 1848–1916; Ger. general 2. Count Helmuth (Karl Bernhard) von, 1800–91; Ger. field marshal; uncle of prec.

mol-to (mōl'tō) *adv.* [It. < L. *multum*, much] *Musica* very; much; used in musical directions

Mo-luc-cas (mōluk'ās) group of islands constituting a province of Indonesia, between Celebes & New Guinea;

c. 32,000 sq. mi.; pop. 790,000; also Molucca Islands

mol. wt. molecular weight

mo-ly' (mō'lē) *n.* [L. < Gr. *mōly*] 1. Classical Myth, an herb of magic powers, as, in Homer's *Odyssey*, that given to Odysseus to protect him from Circe's incantation 2. a European wild garlic (*Allium moly*)

mol-y' (māl'ē) *n.* *dipped form of MOLYBDENUM*

mo-lyb-date (mōlīb'dāt) *n.* a salt of molybdic acid

mo-lyb-de-nite (mōlīb'dā nīt') *n.* native molybdenum sulfide, *MoS₂*, a scaly or foliated, lead-gray mineral, the chief ore of molybdenum

mo-lyb-de-num (-nəm) *n.* [ModL. < *molybdēna*, a lead ore, molybdenite, altered < L. *molybdaena*, lead, galena < Gr. *molybdaina* < *molybdos*, lead] a soft, lustrous, silver-white metallic chemical element; used in alloys, windings for electrical resistance furnaces, points for spark plugs, etc.; symbol, *Mo*; at. wt., 95.94; at. no., 42; sp. gr., 10.2; melt. pt., 2620°C; sublimes at 4507°C

mo-lyb-dic (mōlīb'dik) *adj.* *Chem.* designating or of compounds in which molybdenum has a higher valence (usually 3 or 6) than in the corresponding molybdous compounds

mo-lyb-dous (-dās) *adj.* *Chem.* designating or of compounds in which molybdenum has a lower valence than in the corresponding molybdic compounds

mom (mām) *n.* [Colloq.] mother

MOMA Museum of Modern Art

mom and pop store (stand, etc.) a small retail business, typically family operated and now often franchised

Mom-ba-sa (mām bā'sā, -bas'ā) seaport on the SE coast of Kenya; pop. 130,000

mom-me (mōm) *n.* [*< ?*] [Archaic] a blockhead; fool

mo-ment (mō'mənt) *n.* [ME. < L. *momentum*, movement, impulse, brief space of time, importance < *movimentum* < *movere*, to move] 1. an indefinitely brief period of time; instant 2. a definite point in time or in a series of events 3. a brief time of being important or outstanding 4. importance; consequence (news of great *moment*) 5. *Mech.* a) the tendency to cause rotation about a point or axis b) a measure of this tendency c) the product of a (specified) force, mass, volume, etc. and its perpendicular distance from its axis, fulcrum, or plane 6. *Philos.* any constituent element of a complex entity — *SYN.* see *IMPORTANCE* — *the moment* the present or the immediate future

mo-men-tar-i-ly (mō'mən'ter'ē) *adv.* 1. for a moment or short time 2. in an instant 3. from moment to moment; at any moment

mo-men-tar-y (mō'mən'ter'ē) *adj.* [L. *momentarius*] 1. lasting for only a moment; passing; transitory 2. [Now Rare] recurring every moment; constant 3. likely to occur at any moment — *SYN.* see *TRANSIENT* — *mo-men-tar-i-ness* *n.*

mo-men-t-ly (mō'mənt'ē) *adv.* 1. from instant to instant; every moment 2. at any moment 3. for a single moment

mo-men-to (mō'men'tō) *n.* *erroneous sp.* of *MEMENTO*

moment of truth 1. the point in a bullfight when the matador faces the bull for the kill 2. a critical moment or time that tests and reveals one's true self or makes one face the truth

mo-men-tous (mō'men'təs) *adj.* of great moment; very important [*a momentous decision*] — *mo-men-tous-ly* *adv.* — *mo-men-tous-ness* *n.*

mo-men-tum (mō'men'təm) *n.* *pl.* -tums, -ta (tə) [ModL. < L.; see *MOMENT*] 1. the impetus of a moving object 2. strength or force that keeps growing [*a campaign that gained momentum*] 3. *Physics & Mech.* the quantity of motion of a moving object, equal to the product of its mass and its velocity

mom-ism (mām'iz'm) *n.* [coined (1942) by Philip Wylie, U.S. writer; cf. *MOM & ISM*] excessive devotion to mothers that gives them a dominating role over their children

Momm-sen (mōm'sən; E. mām's'n, -z'n) Theodor (tā'dōr) 1817–1903; Ger. historian

mom-my (mām'ē) *n.* *pl.* -mies *child's term* for *MOTHER*

Mo-mus (mō'məs) [L. < Gr. *Mōmos*, lit. blame, ridicule] Gr. Myth, the god of mockery and censure — *n.* a fault-finder or caviling critic

Mon (mōn) *n.* 1. *pl.* Mons, Mon any member of a people living in Burma east of Rangoon 2. their Mon-Khmer language

mon (mān) *n.* Scot. & North Eng. var. of *MAN*

mon- (mān) same as *MONO-*; used before a vowel

Mon. 1. Monastery 2. Monday 3. Monsignor

mon. 1. monastery 2. monetary

Mo-na (mō'nā) [Ir. *Muadhna*; dim. of *muadh*, noble] a feminine name

mon-a-chal (mān'ā k'āl) *adj.* [ML.(Ec.) *monachalis* < LL.(Ec.) *monachus*; see *MONK*] same as *MONASTIC* — *mon-a-chism* (-kiz'm) *n.*

mon-ac-id (mān'ās'id) *adj.* *n.* same as *MONOACID*

Mon-a-co (mān'ā kō, mā nā'kō) independent principality on the Mediterranean; enclave in SE France; 1/2 sq. mi.; pop. 23,000

mo-nad (mō'nad, mān'ad) *n.* [LL. *monas* (gen. *monadis*) < Gr. *monas* (gen. *monados*), a unit, unity < *monos*, alone; see *MONO-*] 1. a unit; something simple and indivisible 2. *Biol.* a) any simple, single-celled organism, specif. a simple type of flagellated protozoan or protist b) any of the four nuclei formed at the completion of

submittable

fat. äpe, cär; ten. även; is. bite; gē
e for a in ago, e in agent, i in sanity.
ū, Fr. duc; r, Fr. cri; H, G. ich;

apparent to the investigator as he gains facility using the diagrams.

Denoting configurational average by a subscripted angular bracket, we have

$$\langle \langle E^2 \rangle \rangle_c = N^{-1} \sum_{i=1}^N \langle H_{ii}^2 \rangle_c + 2 \langle H_{ii} H_{i,i+1}^2 \rangle_c + 2 \langle H_{ii} H_{i,i-1}^2 \rangle_c + \langle H_{i+1,i+1} H_{i,i+1}^2 \rangle_c + \langle H_{i-1,i-1} H_{i,i-1}^2 \rangle_c \quad (5A)$$

We may eliminate the summation and the factor N^{-1} since terms whose indices can be made identical by replacing i by $i+j$ are equal to each other. Strictly speaking, this means that we are neglecting end effects

and are considering an infinite chain of square wells. With this assumption, we may write

$$\langle \langle E^2 \rangle \rangle_c = \langle H_{ii}^2 \rangle_c + 2 \langle H_{ii} H_{i,i+1}^2 \rangle_c + 2 \langle H_{ii} H_{i,i-1}^2 \rangle_c + \langle H_{i+1,i+1} H_{i,i+1}^2 \rangle_c + \langle H_{i-1,i-1} H_{i,i-1}^2 \rangle_c \quad (6A)$$

By the same argument we could combine, for instance, terms two and five, although this simplification is not used in Eq. (27). However, terms two and three are not identical, nor is

$$\langle H_{ii} H_{i,i+1}^2 \rangle_c = \langle \langle H_{ii} \rangle \rangle_c \langle \langle H_{i,i+1}^2 \rangle \rangle_c$$

These latter points can be proved by direct computation using the probability distribution functions (25).

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Solubility of Carbon in Silicon and Germanium

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The solubility of carbon in silicon has been measured over the temperature range 1560 to 2900°C. The enthalpy of solution is 59 ± 3 kcal/mole. A phase diagram for the system Si—C is presented, embodying these solubility data as well as the results of other high-temperature experiments with silicon carbide. It is found that SiC possesses a peritectic point at $2830 \pm 40^\circ\text{C}$. These studies were carried out in argon at pressures as high as 35 atmos. Solubilities of carbon in germanium were measured in the temperature range 2780 to 3170°C, at argon pressures up to 35 atmos, and a tentative phase diagram is given.

EXPERIMENTS WITH SILICON

Introduction

NEITHER the solubility of carbon in silicon at high temperatures, nor the phase diagram of the system Si—C is known. Nowotny *et al.*,¹ in working with the ternary system Mo—Si—C, proposed two possible phase diagrams for the system Si—C. These were for pressures less than one atmosphere and were based on a limited number of experimental points so that a decision between the two could not be made. The solubility of C in liquid Si has been measured by Hall² up to 1710°C. Others³ have found Si as an impurity in natural diamond, although the conditions of formation are of course unknown. We present here some solubility measurements in liquid Si up to 2900°C, and a proposed phase diagram for the system.

Experimental Procedure

The experiments were carried out in a 50-kw graphite resistance furnace heated by 20 v maximum, 60-cy

ac. This was enclosed in a water-cooled pressure vessel. Argon at a pressure of 35 atmos was used to reduce the tendency of the silicon to evaporate. The vapor pressure of silicon is about one atmosphere at 2800°C over both Si⁴ and SiC.⁵ Figure 1 shows the arrangement of the silicon container, heater, and shields which were used above 2000°C. The entire assembly was of graphite and was supported from the current terminals. On the basis of melting point measurements on iridium wires, the temperature variation over the container was believed to be not over $\pm 25^\circ\text{C}$ from the value at the point of measurement. The mounting of the container within the heater tube was such that no current passed through the Si charge. In this way, changes in the state of the Si, and hence in its electrical resistivity, did not affect the temperature distribution.

The Si was du Pont hyperpure grade, ground with a Pyrex mortar and pestle, screened, and leached with HF to remove any glass contamination in the Si from the mortar. The graphite container was filled with 100 mesh Si (about 0.5 g) and closed with a graphite end plug. No carbon was added; the carbon which was

¹ Nowotny, Parthe, Kieffer, and Benesovsky, *Monatsh. Chem.* 85, 255 (1954).

² R. N. Hall, *J. Appl. Phys.* 29, 914 (1958).

³ F. G. Chesley, *Am. Mineralogist* 27, 20 (1942); F. A. Raal, *ibid.* 42, 354 (1957).

⁴ R. E. Honig, *J. Chem. Phys.* 22, 1610 (1954).

⁵ Drowart, de Maria, and Ingrham, *J. Chem. Phys.* 29, 1015 (1958).

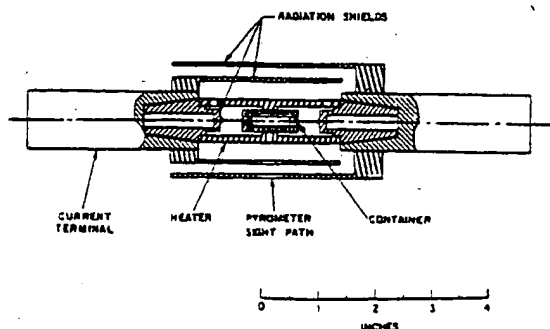


FIG. 1. Graphite resistance furnace (axial cross section).

taken into solution by the liquid silicon came from the walls of the container.

After assembly of the furnace, the pressure vessel was evacuated; it was then refilled twice with argon to a pressure of 20 atmos, vented to atmospheric pressure each time, and finally filled to the operating pressure. A manostat maintained the pressure within ± 1 atmos.

The length of time that the charge was kept at the operating temperature was a compromise. A long time ensured equilibrium, but for long times the Si loss became excessive with graphite containers. Usually

temperature was kept constant at the desired value for periods of one-half to two minutes. No dependence of the C solubility upon the duration of run was observed. The charge was cooled as rapidly as possible after the run by simply shutting off the power to the furnace. Initial cooling rates of $50^\circ\text{C}/\text{sec}$ were typical.

Analysis

The carbon which was in solution in the liquid silicon at high temperatures crystallizes as SiC upon cooling. The Si slug containing the SiC was recovered by oxidizing away the graphite reaction vessel at 1100°C in an O_2 atmosphere. Less than 0.2 mg of SiO_2 was produced by this treatment. The original charge of Si could be accounted for in four portions:

1. Some had reacted with the container, forming a microcrystalline mass of SiC within the pores of the graphite container. Use of a dense graphite ($1.90 \text{ g}/\text{cm}^3$)* for the containers was necessary to avoid excessive losses in this way. This loss occurred mainly during the furnace warmup after the silicon liquified, and in unfavorable cases amounted to over 50 percent of the initial charge.

2. Some remained within the container as elemental Si, frozen into a small slug. This generally ranged from 10 percent to 60 percent of the initial charge of 500 mg, depending on the porosity of the container.

Some precipitated as SiC during cooling of the furnace. Most of the SiC was enclosed within the frozen

mass of Si, although some may have segregated to the surface of the Si. No free C remained within the piece of silicon, as determined both by microscopic examination and by chemical analysis of the residue which remained after the silicon was removed by etching.

4. Little if any Si was lost by evaporation because of the blanketing effect of the argon and the low porosity of the graphite container.

The slug of elemental Si containing the precipitated silicon carbide was lightly sandblasted to remove SiC which was formed in the walls of the graphite container and adhered to the surface of the slug. A thin layer of silicon (less than 25μ) was removed from the slug during the sandblasting. After the sandblasted slug was weighed, the Si was etched away in a mixture of 1 part HF to 3 parts HNO_3 . The SiC residue, which was not attacked by the etch, was dried and weighed. The carbon solubility at the operating temperature of the furnace was computed from the ratio of the mass of SiC to the mass of Si in the slug.

The major source of error in this procedure was the separation of the SiC formed by surface reaction with the container from that formed by reaction with dissolved carbon. It is felt that the experiments done in graphite gave a lower limit to the solubility of C in Si, because some dissolved SiC which was segregated to the surface during cooling may have been lost in sandblasting. Because only a thin layer was removed it is believed that at least 90 percent of the SiC is retained in the slug. Also, from the reproducibility of experiments one concludes that this loss was not serious.

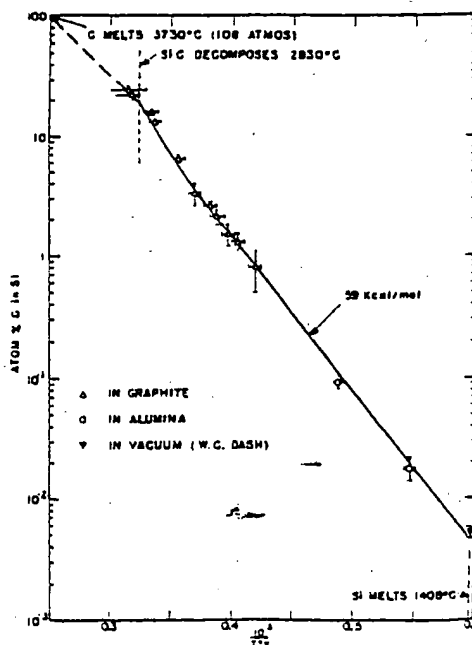


FIG. 2. The solubility of C in Si as a function of temperature.

Two of the measurements were made at 2000°C with different heaters and this change was necessary because a change in the amount of carbon (10–20 g) was needed. In the latter case, SiC was included in the measurement to determine the content of the Si melt. SiC which was dissolved. The total amount of SiC was 5 mg. These experiments were in order to provide a check on the data in graphite. These data were because all the SiC was considered to have gone for redeposition of SiC or have occurred during the measurements performed in graphite is. A measurement of the SiC was made by W. C. Dash using a pedestal clamped at the bottom of a portion melted. The SiC of about 4.1 mg was melted and sank until it reached the bottom. After about 10 minutes, which surrounded the etching and again the weight loss and the measurement of the solubility to be found. silicon at the melting p-

Temperature

All temperature measurements were made with Leeds & Northrup thermocouples which had been recently calibrated. Errors are known to within 1% on the temperature (L) scale. Corresponding scales have at most a 1% transmission through the furnace. Measurements were made separately inside the furnace as a check on the temperature correction to be made. In operation the surface of the furnace was at the bottom of a hole in the furnace. The rough walls of the furnace were painted to a blackbody. The wall of the Si container was at the bottom of the container which had a hole. This hole was so positioned

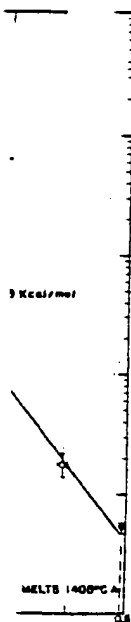
* We are indebted to R. L. Shepard, of the National Carbon Company Research Laboratories, for some samples of a suitable high-purity graphite.

have segregated to the end within the piece of microscopic examination residue which remained etching.

vapor because of and the low porosity

ing the precipitated asted to remove SiC he graphite container slug. A thin layer of oved from the slug he sandblasted slug vay in a mixture of residue, which was d and weighed. The temperature of the atio of the mass of

procedure was the surface reaction with reaction with dis- experiments done in olubility of C in Si, as segregated to the been lost in sand- was removed it is the SiC is retained ucibility of experi- as not serious.



on of temperature.

Two of the measurements at temperatures below 2000°C were made in the same furnace, but with a different heater and an Al_2O_3 container (see Fig. 2). This change was necessary because a larger melt (10–20 g) was needed to dissolve a measurable amount of carbon. In the latter experiments, a weighed crystal of SiC was included in the melt, and its weight loss was used to determine the solubility. As a check, the SiC content of the Si melt was also found. About 2/3 of the SiC which was dissolved from the crystal was recovered. The total amount of SiC lost from the crystal was 5–10 mg. These experiments in ceramic containers were done in order to provide a comparison with those carried out in graphite. These data are upper limits to the solubility, because all the SiC lost from the weighed piece was considered to have gone into solution even though some redeposition of SiC on the walls of the container may have occurred during the run. The agreement between the measurements performed in Al_2O_3 and those performed in graphite is good, as can be seen from Fig. 2. A measurement of the solubility at the Si melting point was made by W. C. Dash. This was performed in vacuum using a pedestal technique.⁴ A vertical bar of Si, clamped at the bottom, was heated at the top until a portion melted. Then a carefully weighed crystal of SiC of about 4.1 mg was inserted from above into the melt and sank until it rested at the liquid-solid interface. After about 10 min the melt was refrozen. The Si which surrounded the SiC crystal was removed by etching and again the SiC crystal was weighed. The weight loss and the mass of Si in the melt enabled the solubility to be found. The carbon solubility in liquid silicon at the melting point is $3.0 \pm 0.3 \times 10^{-18}$ A/cm³.

Temperature Measurement

All temperature measurements were made with a Leeds & Northrup type 8622-C optical pyrometer which had been recently standardized. Calibration errors are known to within $\pm 2^\circ\text{C}$ on the low-temperature (L) scale. Correspondingly the higher temperature scales have at most a $\pm 10^\circ\text{C}$ error. Measurements of transmission through the sight tube window of the furnace were made separately using a tungsten strip lamp inside the furnace as a source, with the window alternately in place and removed. This allowed an accurate correction to be made for transmission losses. During operation the surface of the Si container was visible at the bottom of a hole in the heater tube two diameters deep. The rough walls of the hole and the high emissivity of graphite both tended to make the hole equivalent to a blackbody. The temperature drop across the wall of the Si container was found as a function of temperature by direct measurement on an empty container which had a hole through to the interior. This hole was so positioned that both internal and

⁴ W. C. Dash, J. Appl. Phys. 29, 736 (1958).

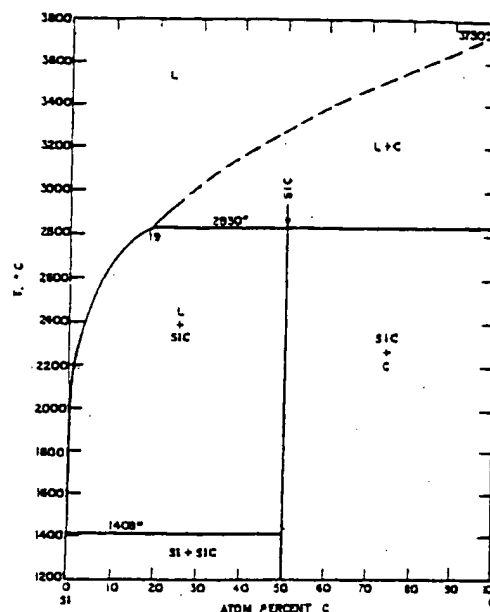


FIG. 3. Phase diagram of the system Si-C for pressures above 108 atmos.

surface temperatures could be measured at the same time. A useful check on the pyrometer was obtained from a plot of heater current vs absolute temperature squared. Above about 1700°C this plot was linear, indicating that radiation was by far the most important mechanism of energy loss.

As a final test, the melting points of Ir (2454°C) and Mo (2622°C) were measured, care being taken to avoid contact between the metal and carbon. We obtained 2446°C and 2634°C, respectively.

Pyrometry errors may be estimated as follows: setting and calibration errors on the instrument, $\pm 10^\circ\text{C}$; window transmission uncertainty, $\pm 20^\circ\text{C}$; and error in the temperature drop across the reaction container wall, $\pm 10^\circ\text{C}$.

Results

The solubility of C in Si as a function of temperature is given in Fig. 2. Some curvature may be noted at concentrations of five atom percent and more, presumably because the solution is nonideal. We find the solubility to be about 1/5 of that reported by Hall.² Recent experiments by J. H. Racette⁷ of this laboratory, in a fused quartz apparatus similar to that used by Hall, indicate that this discrepancy was most likely due to the presence of sufficient oxygen in the melt in Hall's experiment to remove much of the dissolved carbon as CO or CO_2 . The oxygen presumably came from the quartz (SiO_2). In that case the carbon solubility would have appeared to be too high because the SiC crystal placed in the melt continuously lost carbon. Racette's

⁷ J. H. Racette (private communication).

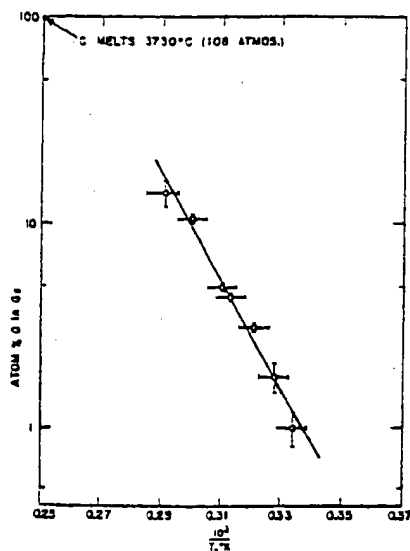


FIG. 4. The solubility of C in Ge as a function of temperature.

experiments show an apparent increase in solubility as the duration of the experiment is increased.

The linear portion of the curve below 1A% carbon has a slope corresponding to an enthalpy of solution of 9 kcal/mole, as calculated from the expression

$$x = \exp\{(\Delta H/R)[(1/T) - (1/T_m)]\}. \quad (1)$$

In this equation, x is the fractional atomic concentration of carbon in silicon at temperature T , ΔH is the enthalpy of solution, R is the gas constant per mole, and T_m is the temperature intercept at unit carbon fraction obtained by extrapolation of the linear portion of the solubility curve. Depending on how the line is drawn through the data points, ΔH may vary ± 3 kcal/mole.

In the course of these experiments it seemed worthwhile to explore as much of the phase diagram of the system Si-C as was accessible. Attempts were made to melt SiC in graphite containers similar to those used for the solubility experiments. The ambient pressure was well in excess of the decomposition pressure in all experiments. A transformation of some originally light green, type 6H, hexagonal, 120-mesh, SiC grain to a markedly different structure occurred at temperatures of $2830 \pm 40^\circ\text{C}$ and above. The SiC found after the transformation was cubic, as analyzed by x-rays. Heating to lower temperatures resulted only in a sintering together of the grains without any gross structural change. This experiment was repeated with cubic SiC grain as the initial charge. It decomposed at the same temperature, and the SiC found after the transformation was also cubic. From this, and from evidence gained from other experiments with SiC at elevated temperatures, we conclude that SiC decomposes without congruent melting, resulting in a silicon-rich liquid in

equilibrium with graphite at temperatures from 2830°C to at least 3160°C . The liquid composition at this latter temperature is approximately 50 atom percent carbon. When the liquid resulting from the decomposition of SiC is cooled below the decomposition temperature, the Si reacts with the dissolved and suspended carbon to form the cubic SiC found by x-ray analysis.

Experiments have also been performed in which cubic SiC grains have been heated in graphite tubes in this furnace to temperatures slightly below 2830°C . At the end of a run lasting several minutes the original cubic SiC is still present although slightly sintered together. Other experiments have been conducted in this apparatus in which both cubic and hexagonal crystals of SiC have been grown side by side from the vapor phase at temperatures of 2600°C . These growing experiments lasted several hours. It is therefore concluded that cubic SiC is stable at these temperatures and does not readily transform into hexagonal SiC in contrast to the conclusions of Baumann.¹ However, it should be noted that the conditions in the present experiments are not the same as those found in commercial SiC furnaces.

Difficulty was encountered in attempting to extend the solubility measurements above 3000°C . Even the high-density graphite containers would not hold liquid silicon at these temperatures. Furthermore the interpretation of the chemical analysis became difficult because the sample after cooling to room temperature usually contained silicon carbide, silicon, and carbon in a carbon container.

The phase diagram shown in Fig. 3 incorporates the

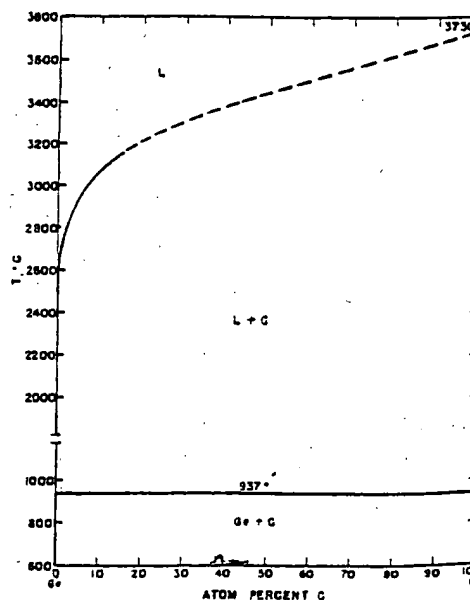


FIG. 5. Tentative phase diagram of the system Ge-C for pressures above 108 atmos.

¹ H. N. Baumann, J. Electrochem. Soc. 99, 109 (1952).

findings described given by Basset,² and by Pell.¹⁰ It confirms the proposals by Hansen.¹¹ Note that it is assumed to be high and are always present. pure carbon at 3730°C over carbon at the required pressure: 15 atmos a temperature reached.⁴

EXPERIMENTAL

The solubility of carbon in germanium has apparently not been previously shown in Fig. 3. The containers to temperatures and have analyzed the limit of 3170°C was germanium evaporation maximum argon pressure below 2780°C the available limited volume of liquid measure accurately. refined; the container scopic graphite with a

The samples were graphite containers. to the graphite, and After weighing, the sample part HF in three parts precipitated graphite

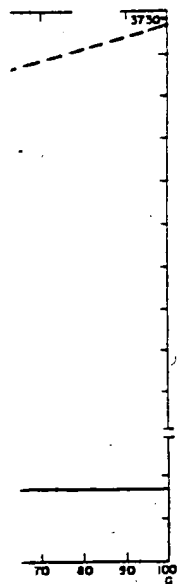
² J. Basset, J. phys. radiat.
¹⁰ E. M. Pell, J. Phys. Chem.
¹¹ M. Hansen, Constitution, p. 378.

temperatures from 2830°C composition at this latter atom percent carbon. the decomposition of position temperature, and studied carbon ray analysis.

formed in which cubic graphite tubes in this below 2830°C. At the times the original cubic tly sintered together. lucted in this appa- gonal crystals of SiC n the vapor phase at growing experiments concluded that cubic and does not readily contrast to the con- should be noted that eriments are not the SiC furnaces.

attempting to extend e 3000°C. Even the ould not hold liquid hermore the interpre- ame difficult because temperature usually id carbon in a carbon

g. 3 incorporates the



system Ge-C for pres-

99, 109 (1952).

findings described above, the melting point of carbon given by Basset,⁹ and the melting point of silicon given by Pell.¹⁰ It confirms the general features of the diagrams proposed by Nowotny *et al.*¹ and quoted by Hansen.¹¹ Note that in Fig. 3 the ambient pressure is assumed to be high enough that solid or liquid phases are always present. This will be at least 108 atmos for pure carbon at 3730°C, since this is the vapor pressure over carbon at the triple point.⁹ At lower temperatures the required pressure will be less. With a pressure of 35 atmos a temperature of about 3400°C can be reached.⁴

EXPERIMENTS WITH GERMANIUM

The solubility of carbon in germanium has apparently not been previously reported. Using the furnace shown in Fig. 1, we have heated Ge in graphite containers to temperatures between 2780°C and 3170°C and have analyzed the results. The upper temperature limit of 3170°C was set by the requirement that the germanium evaporation rate be kept low with a maximum argon pressure of 55 atmos. At temperatures below 2780°C the amount of carbon dissolved in the limited volume of liquid germanium was too small to measure accurately. The Ge had been previously zone refined; the containers were machined from spectroscopic graphite with a density of 1.7 g/cm³.

The samples were recovered by cracking apart the graphite containers. The germanium did not adhere to the graphite, and was in the form of a shiny slug. After weighing, the slug was etched in a mixture of one part HF in three parts HNO₃. The residue was partly precipitated graphite in the form of small flakes and

partly Ge. Apparently the etch was unable to leach all the Ge from the graphite flakes because of the hydrophobic nature of graphite. The residue was therefore roasted in Cl₂ at 1000°C in a quartz crucible to remove the Ge by the formation of volatile GeCl₄. After the remainder of the residue was weighed, it was then burned in O₂ at 1000°C to form CO₂. No evidence of GeO or GeO₂ was found after the oxidation, indicating that the residual Ge had indeed been removed from the graphite.

The results are shown in Fig. 4. If the solubility line were extrapolated to the melting point of Ge, the concentration of C in the liquid would be about 10⁸ atoms per cm³. This is probably a lower limit. The carbon-germanium system is nonideal and such an extrapolation is inaccurate. This extrapolated solubility is a factor of about 10¹⁰ less than the carbon concentration in liquid silicon at its melting point. No evidence of compound formation between carbon and germanium was found up to temperatures of 3170°C. The C-Ge phase diagram therefore appears to be quite simple, and a tentative sketch is shown in Fig. 5. A simple eutectic point probably exists very slightly below the melting point of Ge at 937°C.¹² This is qualitatively similar to the C-Pb and C-Sn systems as deduced from the solubility studies of C in Pb and in Sn at temperatures up to 2300°C.¹³

ACKNOWLEDGMENTS

We are much indebted to W. C. Dash for his measurement of the solubility of C in Si at the Si melting point. Also, we wish to thank R. N. Hall and J. H. Racette for many helpful discussions.

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THE Si-C AND Ge-C PHASE DIAGRAMS*

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Abstract—The solubility of carbon in silicon has been measured over the temperature range 1408°C to 2900°C. The enthalpy of solution is 59 ± 3 kcal/mol. A phase diagram for the system Si-C is presented, embodying these solubility data as well as the results of other high temperature experiments with silicon carbide. It is found that SiC possesses a peritectic point at $2830 \pm 40^\circ\text{C}$. These studies were carried out in argon at pressures as high as 35 atm. Solubilities of carbon in germanium were measured in the temperature range 2780°C to 3170°C, at argon pressures up to 55 atm.

In the literature on silicon carbide, one finds reference to the fact that SiC decomposes at high temperatures. The temperature at which this occurs has been variously cited, and values from 2200 °C^{4,5} to 2700 °C^{8,9} may be found. This wide range, and the fact that most, if not all of these studies were done under non-equilibrium conditions, leads to the belief that a true decomposition of bulk SiC is not what has been observed, but instead a dissociation of the surface caused by preferential evaporation of silicon from the SiC. The heat of formation data of Humphrey *et al.*⁷, when extrapolated to higher temperatures, indicate that decomposition may occur near 2600 °C. If SiC could be melted without the occurrence of bulk decomposition, then crystals could be grown from the melt. Even if SiC does not possess a melting point, further knowledge of the phase diagram of the system Si-C would be useful in crystal growing work.

To investigate the behavior of SiC at elevated temperatures and pressures, the furnace shown in Fig. 1 was built. A water-cooled pressure vessel surrounds the graphite furnace assembly seen in the center. The vessel will hold a pressure of as much as 1000 lb/in.²; this dense atmosphere retards the evaporation of silicon which would otherwise be quite troublesome.

Figure 2 shows a cross-section of the furnace assembly. All the parts are made of graphite. The two current terminals support a tubular heater, two radiation shields, and a container which was used in the experiments on the solubility of carbon in silicon. The container is supported in such a way that it carries no current. About 50 kW is available to heat this assembly, enabling temperatures of about 3500 °C to be reached.

Optical pyrometry is the only reasonably accurate way of measuring temperature above 1900 °C, which is the limit for platinum-alloy thermocouples. A sight tube and window are provided in the side of the pressure vessel, and openings are left in the radiation shields and heater tube to allow measurement of the internal temperature of the furnace. Corrections to the pyrometer reading must be made for window transmission

* A more complete account of this work has been accepted for publication in the *Journal of Chemical Physics*.

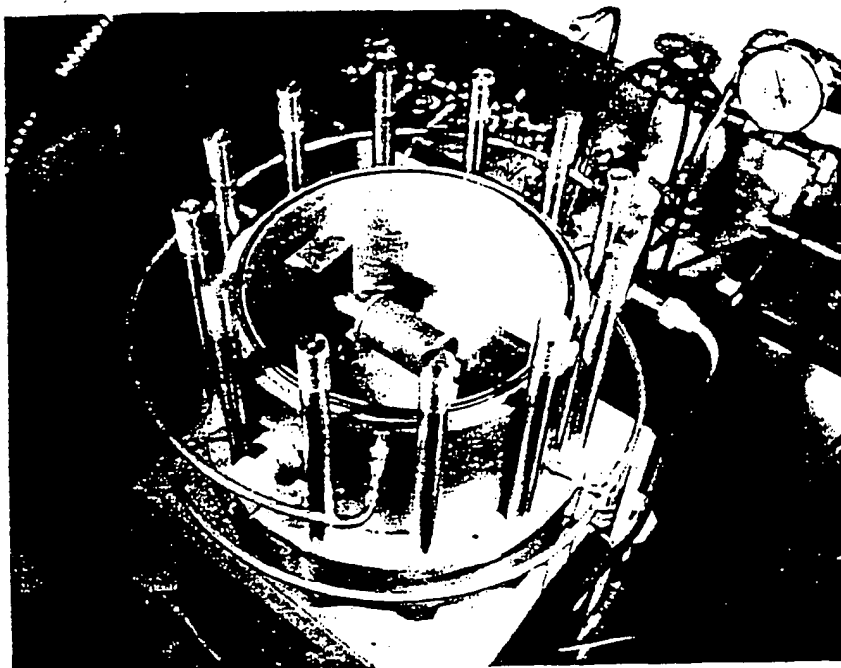


FIG. 1. Interior view of pressure vessel, showing high-temperature furnace in place.

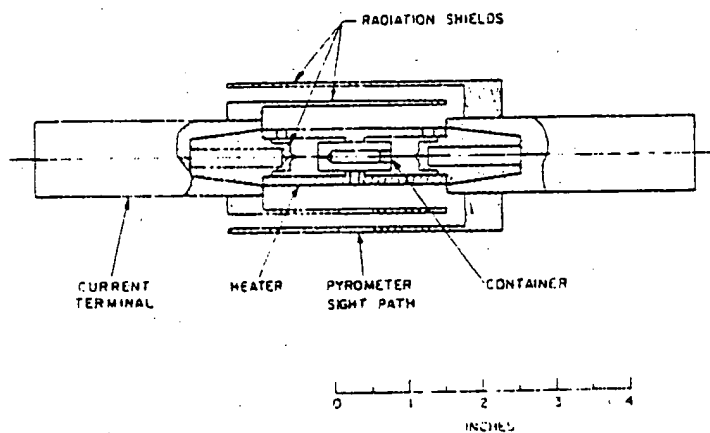


FIG. 2. Cross-section sketch of furnace assembly.

and the emissive properties of the surface whose temperature is to be found. As a check, the melting points of Ir and Mo were measured. The temperatures found were within

$\pm 10^\circ \text{C}$ of the published values. Under ordinary circumstances, temperature measurements are believed to be within $\pm 30^\circ \text{C}$ of the true value except at the very highest temperatures (say 3000°C or more) where errors may amount to 50°C .

To determine the solubility of C in Si, the inner container was filled with Si. The container was sealed with a tight fitting plug, assembled into the furnace and heated to some temperature between 2100 °C to 2900 °C. Thermal equilibrium was achieved quite rapidly, and the sample was held at a fixed temperature for one or two minutes, after which the power was shut off. Then the furnace temperature decreased rapidly by radiation cooling and the Si inside froze. The graphite container was removed from the furnace and burned away from the Si by heating in O_2 . The original charge of Si can be accounted for as follows:

1. Some had reacted with the container, forming a micro-crystalline mass of SiC within the pores of the graphite.
2. Some remained within the container as elemental Si, frozen into a slug. This ranged from 10 to 60 per cent of the initial charge of 500 mg.
3. Some combined with the dissolved C in the liquid Si, and precipitated within the frozen slug of Si as SiC. No free C was found inside the slug of Si.
4. Very little, if any, Si was lost by evaporation because of the blanketing effect of the high pressure argon atmosphere, and the low permeability of the high density graphite.

The loosely adhering SiC, formed by reaction with the container, was carefully removed from the frozen slug. The slug was then weighed, etched in $HF-HNO_3$ to remove the Si, and the remaining SiC found inside the Si slug was weighed. With these data the solubility could be found for the temperature in question.

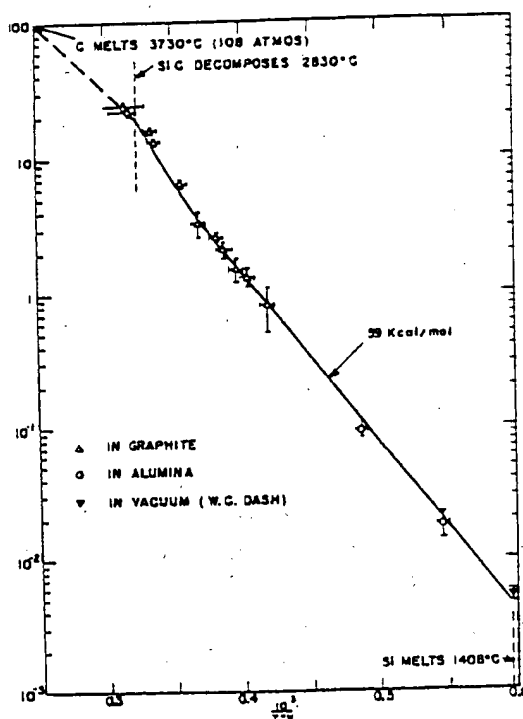


FIG. 3. The solubility of carbon in silicon as a function of reciprocal temperature.

Below 2000 °C the solubility is so low that an accurate determination with 500 mg of Si solvent was impossible. Two runs of 10 and 20 g melts were done in recrystallized alumina crucibles, with a different furnace arrangement in the pressure vessel. Here a weighed single crystal of SiC was included in the melt, and its weight loss combined with the known weight of Si in the melt enabled the solubility to be found.

Finally, Dash² performed a measurement of the solubility at the Si melting point, again by measuring the weight loss of a single crystal of SiC. His melt of Si was supported on top of a pedestal¹ of Si in vacuum. The SiC crystal rested on the liquid-solid interface, so there is little question of temperature measurement accuracy.

These data are all summarized in Fig. 3. The consistency of the results of the various experiments is good. Two points of particular interest are the solubilities of carbon in liquid Si at the Si melting point, 5×10^{-3} atom per cent (5×10^{17} C/cm³ Si), and at the decomposition temperature of 2830 °C. This is the maximum temperature at which SiC could be grown from a Si solution, and the carbon content of the melt here is 19 atom per cent. This high solubility indicates that growth from solution is feasible. The vapor pressure of Si over Si⁸ and over SiC³ at this temperature is of the order of 1 atm and rising rapidly, so the need for a pressurized atmosphere to maintain stable conditions for any length of time is apparent.

Another way of plotting these data is on a temperature vs. composition phase diagram, Fig. 4. The behavior of SiC at the decomposition temperature is more clearly seen. This temperature, 2830 °C, was found by heating samples of SiC grain in containers such as

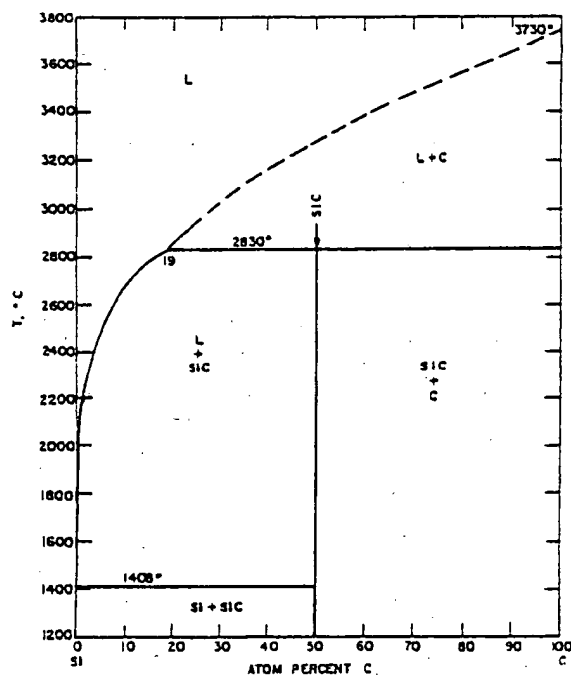


FIG. 4. Phase diagram of the binary system Si-C.

were used for the solubility measurements. All samples whose temperature exceeded this value were decomposed, while none were affected which were cooler. Two fortunate trials, one with hexagonal SiC and one with cubic, were half decomposed and half not, so the temperature was determined as closely as pyrometry errors will allow.

Some measurements have also been performed on the C solubility in Ge, in a manner similar to that used for Si. Figure 5 shows a tentative Ge-C phase diagram based on these measurements. Solubilities of 12 per cent at 3170 °C were reached, where the Ge vapor pressure is several atmospheres. No evidence of compound formation was seen. A very rough extrapolation of these data to the melting point of Ge can be made, indicating that the C solubility in liquid Ge at this temperature is of the order of 10^{10} C/cm³ Ge.

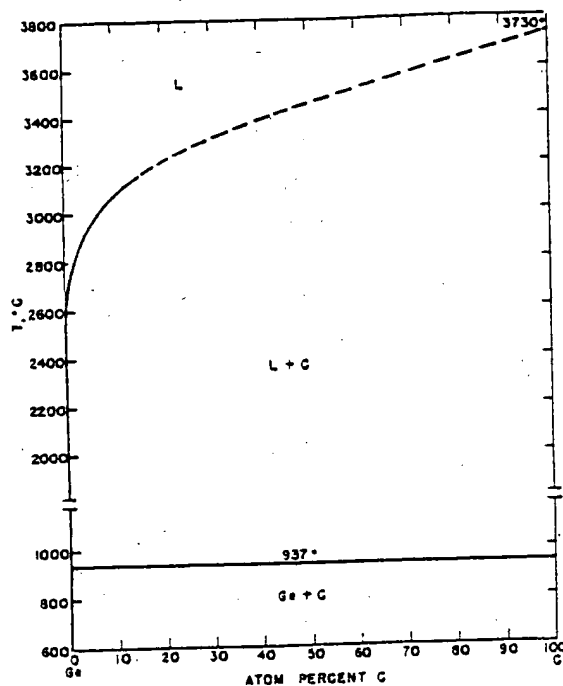


FIG. 5. Tentative phase diagram of the binary system Ge-C.

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